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Lauridsen, K.

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Development of a Model for the Assessment of Radiation Fields Around Nuclear Power Plant Components

by

Kurt Lauridsen

Research Establishment Risø

Reactor Technology Department

Abstract

A method is presented for the calculation of the radioactivity inventories of components in the coolant circuit of a nuclear power plant. The study concentrated on power plants with boiling water reactors of General Electric BWR/6 type. The model accounts for the transport of fission products and activation products between different components, as well as the exchange of material between the interior surfaces of a component and the bulk coolant in this component. A simple point-kernel shielding calculation is applied to evaluate the radiation field outside the components.

CONTENTS

1. INTRODUCTION

During the last decade, in which numerous nuclear power plants have been taken into service and achieved a certain age, the problem of occupational radiation exposure has become a serious one. Many plants have been forced to engage extra staff in order not to exceed the maximum permissible doses. Not only does this constitute a problem of economy - which might not be so hard to overcome - but it might also create a problem of resources. For example, if the radiation fields are allowed to reach such levels that the number of persons required to perform a given job - so nobody receives excess doses - exceeds the number of qualified people available.

Measurements of radiation fields and doses are performed at all nuclear power plants as part of the daily routine, and at many installations additional efforts are made to reveal the most important reasons for the high doses. Findings show that a substantial increase in radiation levels at some plants has been caused by a poor choice of construction materials leading to the build-up of long-lived corrosion product activity, e.g. ^{60}Co. In other cases, inadequate design and layout have hampered maintenance and repair work, and thus personnel doses have reached high values.

These experiences have led to changes in working procedures, as well as changes in the design and the choice of materials where this has been possible. Furthermore, this experience has been utilised in the design of new generations of nuclear power plants.

In the work described in the present report, an attempt has been made to set up a mathematical model by which it should be possible to predict the time-dependent radiation levels at various locations in a nuclear power plant with a boiling water reactor. The input data required by this model are: basic nuclear data, properties of materials, flow-data for the reactor and energy-conversion system, the power history of the reactor, and the geometry of the locations where radiation levels are to be calculated.

The calculation of radiation levels is divided into two main parts: a source-strength evaluation and a shielding calculation. Three types of radiation source are considered: fission

products released from the fuel, corrosion products released from system components and activated by passing through the core, and nuclides created by activation of the coolant itself - often called intrinsic activation products. The model describes the transport of source-nuclides from the reactor through the energy conversion system and back to the reactor taking into consideration radioactive decay, activation, burn-up by neutron capture and the exchange of material with the component-walls by erosion and deposition. Shutdowns of the reactor may occur.

When the source strengths of the various plant components have been found, the attenuation of the radiation as a result of component walls and shielding is calculated. This is done by means of a simple point-kernel method applying build-up factors; the source geometry is assumed to be cylindrical as this is applicable to many power plant components. Cylindrical or planar shields can be treated.

A power plant with a boiling water reactor of the General Electric BWR/6 type was chosen for the study. Two main reasons for this choice were: 1) at the time when the study was initiated (February 1974) a BWR seemed most likely to be chosen for a Danish nuclear power plant, and 2) the BWR/6 was the type for which most information was available [1].

It is stressed that the evaluations and assessments made in the present work are based on normal operation of the power plant. They do not claim to be - and probably are not - valid for accident conditions where failure of fuel and leakage of components may occur.

2. THE POWER PLANT MODEL

The model is intended to be able to give an assessment of the radioactive inventories in components throughout the whole plant. Therefore it must describe the coolant-path in some detail, i.e. it must comprise a relatively large number of components. On the other hand, computer time puts a limit to the degree of detail. The compromise became a 39-component description of the reactor and turbine systems, as shown in fig. 2.1. The coolant path is essentially closed, but there are two locations - apart

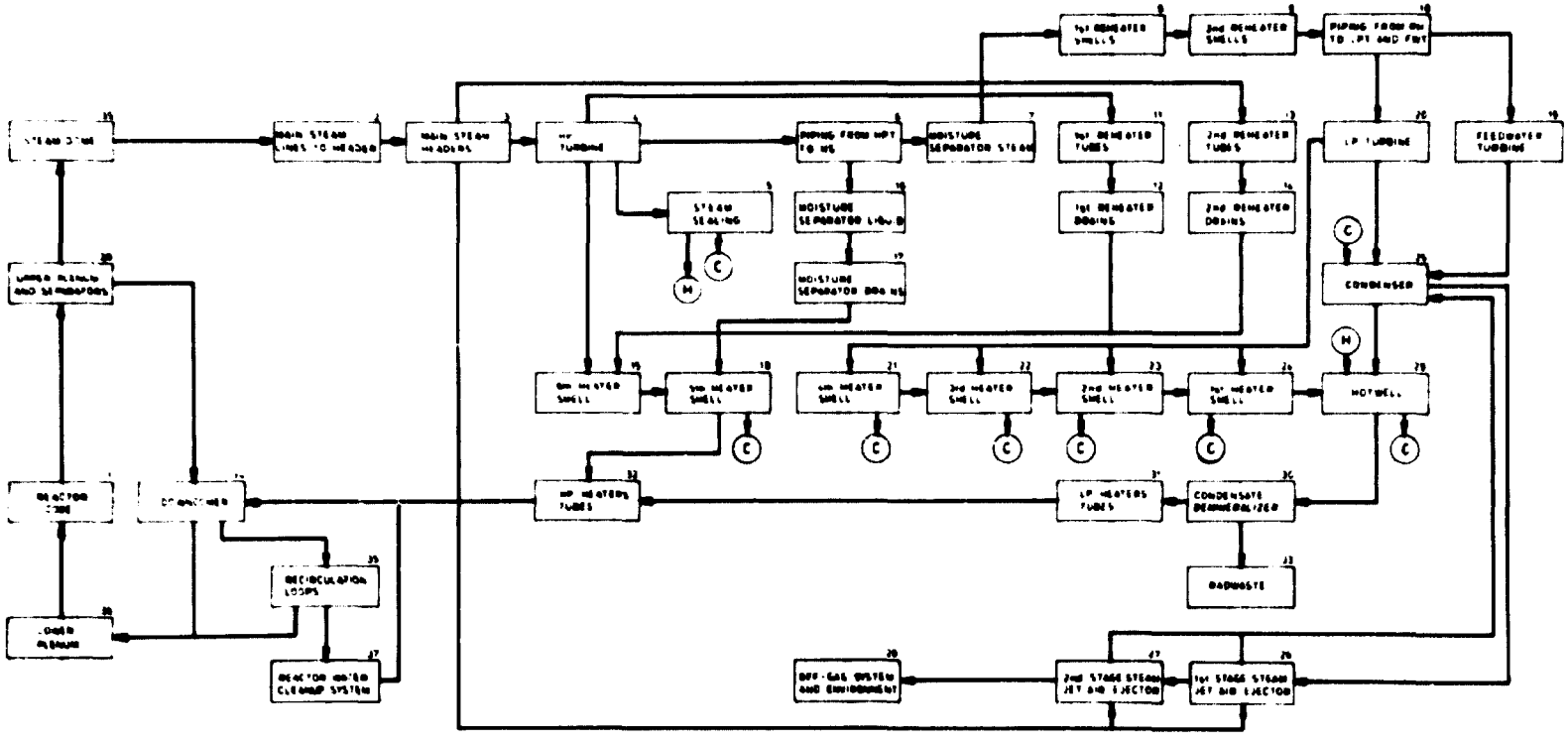

Fig. 2.1. The power plant model.

from leaks - where radioactive material is released, namely to the radwaste and the off-gas systems. These systems are only represented by one "box" each in the model, although they consist of many components. The model is thus unable to predict inventories of single components in these systems. In fact, the values calculated for these two systems only indicate what remains after decay of the activity that has passed through the nozzles to the radwaste and off-gas system. They do not indicate whether this activity is still in the plant, been released to the atmosphere, or dispatched to the waste storage.

The mass-transport between components is described as illustrated in fig. 2.2.

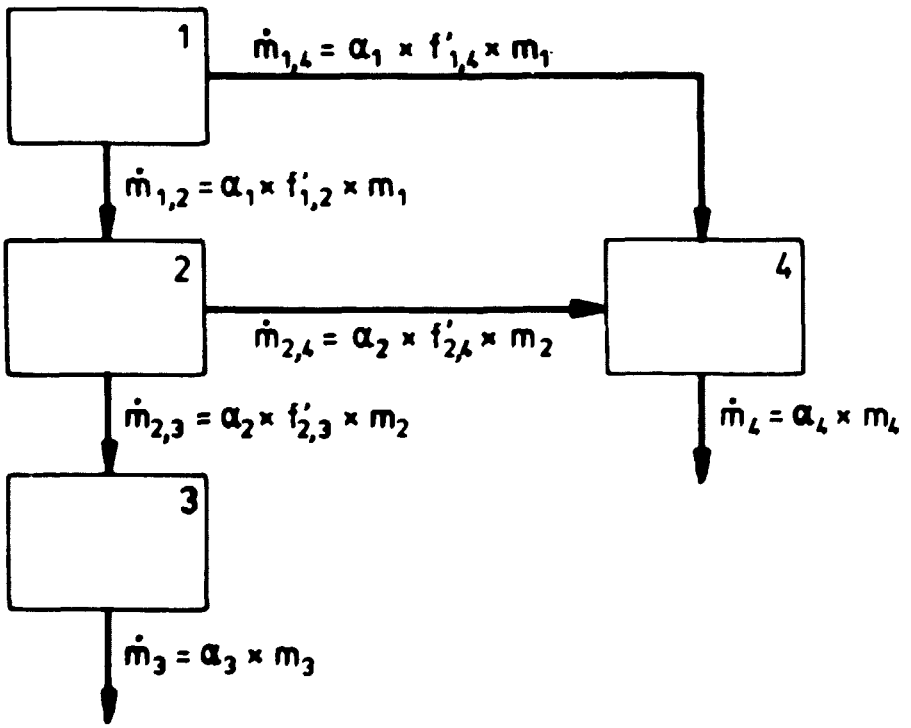

Fig. 2.2. Mass transport description.

Let m_i be the mass of the inventory of component i; the mass flow from component i to k is $\dot{m}_{i,k}$. The total mass flow out of component i is

$$\dot{m}_i = \sum_{k \neq i} \dot{m}_{i,k}.$$

The inventory change rate of component i is then

$$\alpha_i = \frac{\dot{m}_i}{m_i} .$$

A flow-split factor is defined as

$$f'_{i,k} = \frac{\dot{m}_{i,k}}{\dot{m}_i}$$

Using the inventory change rate and the flow split factor, the mass flow from component i to component k can be written as

$$\dot{m}_{i,k} = \alpha_i \, f'_{i,k} \, m_i . \qquad (2\text{-}1)$$

In order to be able to treat nuclear processes such as decay and activation, the mass flow is converted into flow of atoms. For a nuclide j homogeneously mixed with the coolant, we find

$$\dot{n}_{i,k,j} = \frac{N_{Av} \cdot C_{i,k,j}}{M_A} \dot{m}_{i,k} = \frac{N_{Av} \cdot C_{i,k,j}}{M_A} \alpha_i \, f'_{i,k} \, m_i \qquad (2\text{-}2)$$

where

$\dot{n}_{i,k,j}$ = the flow of atoms of nuclide j from component i to component k

N_{Av} = the Avogadro number

M_A = the atom weight of nuclide j, and

$C_{i,k,j}$ = the concentration of nuclide j in the coolant flowing from i to k.

For nuclides which are constituent parts of the coolant, i.e. oxygen and hydrogen, there is no difference in the concentration for different flow-paths. But in components where the steam- and water-phases are separated partly or totally, differences may occur for nuclides dissolved in the coolant. In such cases, noble gases will tend to follow the steam whereas iodine and corrosion products, for instance, mainly join the water phase.

The number of atoms of nuclide j contained in the coolant in component i is:

$$n_{i,j} = \frac{N_{Av}}{M_A} C_{i,j} \cdot m_i \qquad (2\text{-}3)$$

where

$C_{i,j}$ = the (average) concentration of nuclide j in component i.

The concentrations $C_{i,j}$ and $C_{i,k,j}$ are connected by the expression

$$C_{i,j} \dot{m}_i = \sum_k C_{i,k,j} \dot{m}_{i,k} \qquad (2\text{-}4)$$

or

$$C_{i,j} = \sum_k C_{i,k,j} \frac{\dot{m}_{i,k}}{\dot{m}_i} = \sum_k C_{i,k,j} f'_{i,k} \cdot \qquad (2\text{-}5)$$

Equations (2-2) and (2-3) yield:

$$\dot{n}_{i,k,j} = \frac{C_{i,k,j}}{C_{i,j}} \alpha_i f'_{i,k} n_{i,j} \qquad (2\text{-}6)$$

Introducing the partition factor $f_{i,k,j}$ for the flow of nuclide j from component i to k:

$$f_{i,k,j} \equiv \frac{C_{i,k,j}}{C_{i,j}} f'_{i,k} , \qquad (2\text{-}7)$$

equation (2-6) becomes:

$$\dot{n}_{i,k,j} = \alpha_i f_{i,k,j} n_{i,j} . \qquad (2\text{-}8)$$

The main reason for using the partition factors here is that they are available in[1)] for most of the components used in this model. With respect to partition factors, the nuclides are divided into three classes: noble gases, halogens, and others. Accordingly, each component is assigned three partition factors, which, of course, may be equal if, for instance, there is no separation of phases. The partition factors fulfil the condition:

$$\sum_k f_{i,k,j} = 1 \qquad (2\text{-}9)$$

which is easily shown

$$\sum_k f_{i,k,j} = \sum_k \frac{C_{i,k,j}}{C_{i,j}} f'_{i,k} = \frac{1}{C_{i,j}} \sum_k C_{i,k,j} \frac{\dot{m}_{i,k}}{\dot{m}_i}$$

$$= \frac{1}{C_{i,j} \cdot \dot{m}_i} \sum_k C_{i,k,j} \dot{m}_{i,k} = \frac{1}{C_{i,j} \cdot \dot{m}_i} C_{i,j} \dot{m}_i$$

q.e.d.

Equation (2-8) represents a system of equations yielding the inventories of the power plant components, if no processes other than mass transport were active. In the following sections of this report processes such as corrosion and radioactive decay are included.

3. MODEL FOR FISSION PRODUCTS

The description of fission product behaviour is divided into two sections: 1) Normal operation and 2) Shutdowns. Normal operation is here defined as steady-state operation at a fixed power level.

3.1. Normal Operation

For a component in the plant model (fig. 2.1), the changes in the number of atoms of fission product nuclide j are described by the following firstorder, linear differential equation:

$$\frac{dn_{ij}}{dt} = S_{ij}(t) + b_{j-1,j}\lambda_{j-1} n_{i,j-1} + \sum_k \alpha_k f_{k,i,j} n_{k,j} + e_{ij} w_{ij}$$

$$- (\lambda_j + \alpha_i + \sigma_j \phi_i + d_{ij}) n_{ij}. \qquad (3\text{-}1)$$

Index j-1 denotes the precursor of nuclide j in the decay chain. If j=1, terms with index j-1 are ignored.

$S_{ij}(t)$ = release rate (atoms/s) of nuclide j in component i. $S_{ij} \neq 0$ only for component no. 1: Reactor Core, where fission products may be released from the fuel to the coolant.

λ = decay constant (s^{-1})

$b_{j-1,j}$ = branching ratio - the fraction of decays of nuclide j-1 that result in nuclide j.

σ = microscopic absorption cross section (cm^2)

Φ_i = neutron flux in component i ($cm^{-2}\ s^{-1}$), $\Phi_i \neq 0$ only for the reactor core (i=1)

e_{ij} = erosion constant (s^{-1})

d_{ij} = deposition constant (s^{-1})

w_{ij} = number of atoms of nuclide j on the (inside) walls of component i.

3.1.1. Discussion of Equation (3-1)

In this paragraph equation (3-1) is discussed in detail, taking it term by term.

3.1.1.1. Source Term. The only component in which fission products are produced is the reactor core. Accordingly, this component is the only one that may have a source term $S_{ij}(t)$ not equal to zero. Release of fission products to the coolant may occur in two different ways: from "tramp uranium" and through holes in the fuel cladding. "Tramp uranium" means the traces of fissile material left on the outside of the fuel elements after the fabrication process. Usually this contribution is negligible due to thorough cleaning of the fuel elements before insertion in the reactor. The release rate through holes in the cladding is assumed to be proportional to the inventory of the fuel:

$$R_j(t) = D\nu_j\, A_j(t) \qquad (3\text{-}2)$$

$$A_j(t) = G \cdot y_j(1-e^{-\lambda_j t}) \cdot \frac{1}{3.7\cdot 10^{10}} \qquad (3\text{-}3)$$

where:

t = irradiation time (s)

$R_j(t)$ = release rate of nuclide j (Curie/s)

D = fraction of fuel rods with defective cladding

ν_j = escape rate coefficient (s^{-1})

$A_j(t)$ = activity inventory of nuclide j (Curie)

G = fission rate (s^{-1})

Y_j = fission yield of nuclide j (atoms/fission)

Equation (3-3) assumes that fission is the only process producing nuclide j.

The escape rate coefficient, ν_j, was originally deduced by means of diffusion theory to depend on λ_j as $\sqrt{\lambda_j}$[2-4]. Later investigations[1] seem to indicate that another exponent of λ_j would give a better agreement with measured releases. Thus

$$\nu = k_1 \lambda^m \tag{3-4}$$

and accordingly:

$$R_j(t) = k_2\, y_j \lambda_j^m\, (1-e^{-\lambda_j t}), \tag{3-5}$$

where k_1 and k_2 are constants. In eq. (3-5) the exponent m will determine the spectrum of the release, while the constant k_2 will determine the absolute magnitude of the total release. It is anticipated that only noble gases and halogens are released through leaks in the fuel, and in accordance with[1], constants k_2 and m in equation (3-5) are given one set of values for halogens and another set for noble gases. For noble gases, the following expression is used

$$R_j(t) = 26 \cdot y_j \cdot \lambda_j^{0,4}(1-e^{-\lambda_j t}) \quad Ci/s. \tag{3-6}$$

For halogens,

$$R_j(t) = 24 \cdot y_j \cdot \lambda_j^{0.5}(1-e^{-\lambda_j t}) \quad Ci/s. \tag{3-7}$$

The exponents of λ_j were determined by fitting measured data to eq. (3-5), cf. [1]. The values of the constant k_2 were derived from the assumption of a total noble radiogas release of 0.1 Ci/s and a ^{131}I release of 700 μCi/s measured after 30 minutes delay, i.e.

$$\sum_{\substack{\text{Noble}\\ \text{gases}}} R_j(t)\, e^{-\lambda_j \cdot 30\ \text{min}} = 0.1\ \text{Ci/s} \qquad (3\text{-}8)$$

and

$$R_I(t) e^{-\lambda_I \cdot 30\ \text{min}} = 700\ \mu\text{Ci/s} \qquad (3\text{-}9)$$

The 30 minutes delay was chosen because contributions from short-lived nuclides are thus eliminated, and because 30 minutes is the hold-up time before release to the atmosphere of off-gas at several older plants. At newer plants, hold-up times tend to be longer.

To express the source term in atoms/s as it is in eq. (3-1), one must divide by λ_j:

$$S_{ij}(t) = \frac{1}{\lambda_j} R_j(t) \quad \text{for } i = 1. \qquad (3\text{-}10)$$

3.1.1.2. Decay Terms. If nuclide j is a member of a decay chain and has a precursor j-1, some "j-atoms" are produced by the decay of "j-1-atoms", giving the term

$$b_{j-1,j} \lambda_{j-1}\, n_{i,j-1}$$

in equation (3-1). Usually, the branching ratio $b_{j-1,j}$ equals 1, but branching does occur and then $b_{j-1,j}$ becomes <1. Likewise, "j-atoms" disappear by decay giving the term

$$-\lambda_j\, n_{i,j}.$$

In some cases significant amounts of a nuclide are produced by neutron capture in the neighbour-isotope (or perhaps by other processes). These cases are treated by replacing the term

$$b_{j-1,j}\lambda_{j-1}\, n_{i,j-1}$$

by

$$\sigma_p\, \Phi_i\, n_{i,p},$$

where index p denotes the parent nuclide in the capture process.

The most frequently occurring decay chain for fission products consists of a series of nuclides of equal mass undergoing β^- decays. When neutron capture is included, the "decay chain" becomes a combination of two (or more) such decay chains. Figure 3.1 shows an extract of the chart of the nuclides that illustrates this. Taking, for instance, ^{88}Kr, this nuclide may be produced in 3 ways: directly from fission, by β^-decay of ^{88}Br, and by neutron capture in ^{87}Kr. And if one wishes to calculate the inventory of ^{90}Y, the following paths must (theoretically) be considered:

1) direct yield from fission

2) $^{90}Rb \xrightarrow{\beta^-} {}^{90}Sr \xrightarrow{\beta^-} {}^{90}Y$

3) $^{89}Kr \xrightarrow{\beta^-} {}^{89}Rb \xrightarrow{\beta^-} {}^{89}Sr \xrightarrow{n,\gamma} {}^{90}Sr \xrightarrow{\beta^-} {}^{90}Y$

4) $^{88}Br \xrightarrow{\beta^-} {}^{88}Kr \xrightarrow{\beta^-} {}^{88}Rb \xrightarrow{n,\gamma} {}^{89}Rb \xrightarrow{\beta^-} {}^{89}Sr \xrightarrow{n,\gamma} {}^{90}Sr \xrightarrow{\beta^-} {}^{90}Y$

5) $^{88}Br \xrightarrow{\beta^-} {}^{88}Kr \xrightarrow{\beta^-} {}^{88}Rb \xrightarrow{\beta^-} {}^{88}Sr \xrightarrow{n,\gamma} {}^{89}Sr \xrightarrow{n,\gamma} {}^{90}Sr \xrightarrow{\beta^-} {}^{90}Y$

6) $^{88}Sr \xrightarrow{n,\gamma} {}^{89}Sr \xrightarrow{n,\gamma} {}^{90}Sr \xrightarrow{\beta^-} {}^{90}Y$

7) $^{87}Br \xrightarrow{\beta^-} {}^{87}Kr \xrightarrow{n,\gamma} {}^{88}Kr \xrightarrow{\beta^-} {}^{88}Rb \xrightarrow{n,\gamma} {}^{89}Rb \xrightarrow{\beta^-} {}^{89}Sr \xrightarrow{n,\gamma} {}^{90}Sr \xrightarrow{\beta^-} {}^{90}Y$

8) $^{87}Br \xrightarrow{\beta^-} {}^{87}Kr \xrightarrow{\beta^-} {}^{87}Rb \xrightarrow{n,\gamma} {}^{88}Rb \xrightarrow{n,\gamma} {}^{89}Rb \xrightarrow{\beta^-} {}^{89}Sr \xrightarrow{n,\gamma} {}^{90}Sr \xrightarrow{\beta^-} {}^{90}Y$

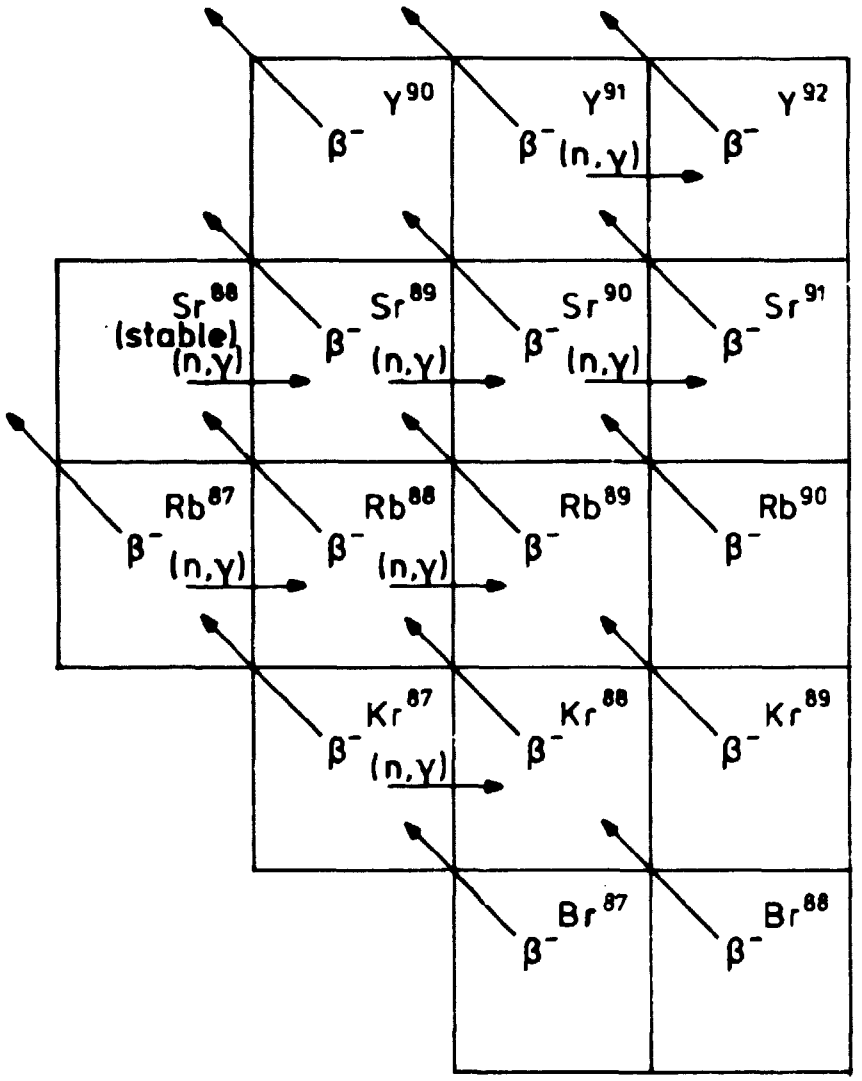

Fig. 3.1. Extract from chart of the nuclides.

9) $^{87}Br \xrightarrow{\beta^-} {}^{87}Kr \xrightarrow{\beta^-} {}^{87}Rb \xrightarrow{n,\gamma} {}^{88}Rb \xrightarrow{\beta^-} {}^{88}Sr \xrightarrow{n,\gamma} {}^{89}Sr \xrightarrow{n,\gamma}$

$^{90}Sr \xrightarrow{\beta^-} {}^{90}Y$

10) $^{87}Br \xrightarrow{\beta^-} {}^{87}Kr \xrightarrow{n,\gamma} {}^{88}Kr \xrightarrow{\beta^-} {}^{88}Rb \xrightarrow{\beta^-} {}^{88}Sr \xrightarrow{n,\gamma} {}^{89}Sr \xrightarrow{n,\gamma}$

$^{90}Sr \xrightarrow{\beta^-} {}^{90}Y$

The contributions from the different pathways must be added together in order to find the inventory of ^{90}Y. Of course, not all the processes specified above are equally important, but it may be diffucult to judge beforehand which ones are most important.

3.1.1.3. Mass Transport Terms. The transport of atoms between the components of the plant is described by the terms

$$\sum_k \alpha_k f_{k,i,j} n_{kj} \text{ and } - \alpha_i n_{ij},$$

which were discussed in detail in chapter 2 of this report.

3.1.1.4. Exchange With Walls. When passing through a component, some atoms may be deposited on the surfaces inside the component, and some that were already deposited may be eroded away from the surfaces. A first-order description of these processes is attempted in eq. (3-1) by introducing the terms

$$e_{ij} w_{ij} \text{ and } - d_{ij} n_{ij},$$

which link the system of equations represented by (3-1) to another system of equations, valid for the inside surfaces of the components:

$$\frac{dw_{ij}}{dt} = d_{ij} n_{ij} + b_{j-1,j} \lambda_{j-1} w_{i,j-1} - (e_{ij} + \lambda_j + \sigma_j \phi_i) w_{ij}. \quad (3\text{-}11)$$

The deposition/release processes are not thought to be important to the activity of the coolant, i.e. n_{ij}, but it is

desirable to assess the values of w_{ij} - the number of atoms deposited on the surfaces - for instance, on turbine parts and other areas where inspection and repairs are expected several times during the plant life. However, constants e_{ij} and d_{ij} are poorly determined, especially for fission products. Models similar to eq. (3-11) have been proposed for corrosion products [5-6], and experiments conducted to determine the constants. Unfortunately, there is little agreement between the results obtained in different measurements. Furthermore, one cannot expect fission products to behave like corrosion products with respect to deposition and release. Presumably noble gases do not adhere to surfaces at all, and halogens may undergo chemical reactions on their way through the system and so change characteristics with respect to deposition and release. Fission products other than those mentioned above are more likely to follow the same pattern as corrosion products. However, it must be stressed that the deposition - and release - terms were included in the model for fission products in order to show the existence of the problem, and no claim is made that the best solution has been found. Hence the results obtained for deposits must not be taken too seriously at present. More experiments and measurements on working power plants are needed before a reliable model can be set up for these phenomena.

3.1.1.5. Neutron Absorption Term. In equation (3-1) the term

$$-\sigma_j \zeta_i n_{ij}$$

describes the removal of nuclide j from component i by neutron absorption. This process takes place mainly in component no. 1: the reactor core, but it also occurs in the downcomer, although to a much lesser extent due to the lower neutron flux. In the computer code it is assumed that absorption only takes place in the core. The product of cross section and neutron flux represents the integrated value over all energies. The computer code does not at present contain facilities for handling multigroup cross sections, so the input value of σ_j must be chosen as a one-group cross section, which can be derived by means of existing reactor

physical codes.

3.1.2. Integration of the Equations.

Equation (3-1) represents a system of NI x NJ linear first--order differential equations, where NI is the number of components in the power plant model and NJ is the number of nuclides in the decay-chain under consideration. NI = 39, and NJ varies between 1 and 7. Equation (3-11) represents another NI x NJ system linked to the first system by the depositon - and release - terms.

The integration method chosen for these systems is a so-called "Modified Euler" by which the value of the variable y at time $t_o + \Delta t$ is expressed as:

$$y(t_o + \Delta t) = y(t_o) + \tfrac{1}{2}\Delta t \left\{ \frac{dy}{dt}\Big|_{t_o} + \frac{dy}{dt}\Big|_{t_o + \Delta t} \right\} \qquad (3\text{-}12)$$

Expressing the time dependence in terms of step number in the numerical integration gives

$$y^{n+1} = y^n + \tfrac{1}{2}\Delta t \left\{ \dot{y}^n + \dot{y}^{n+1} \right\} . \qquad (3\text{-}13)$$

Applying this formula to w_{ij} yields, by means of eq. (3-11),:

$$w_{ij}^{n+1} = w_{ij}^n + \tfrac{1}{2}\Delta t \left\{ d_{ij}(n_{ij}^n + n_{ij}^{n+1}) + b_{j-1,j}\lambda_{j-1}(w_{i,j-1}^n + w_{i,j-1}^{n+1}) \right.$$

$$\left. -(e_{ij} + \lambda_j + \sigma_j \phi_i)(w_{ij}^n + w_{ij}^{n+1}) \right\} \quad \Rightarrow$$

$$\left\{ 1 + \tfrac{1}{2}\Delta t \ (e_{ij} + \lambda_j + \sigma_j \phi_i) \right\} w_{ij}^{n+1} =$$

$$\left\{ 1 - \frac{\Delta t}{2}(e_{ij} + \lambda_j + \sigma_j \phi_i) \right\} w_{ij}^n + \frac{\Delta t}{2} b_{j-1,j} \lambda_{j-1}(w_{i,j-1}^n + w_{i,j-1}^{n+1})$$

$$+ \frac{\Delta t}{2} d_{ij}(n_{ij}^n + n_{ij}^{n+1}) \quad \Rightarrow$$

$$w_{ij}^{n+1} = \frac{1}{1+\frac{\Delta t}{2}\eta_{ij}}\{(1-\frac{\Delta t}{2}\eta_{ij})w_{ij}^{n} + \frac{\Delta t}{2} b_{j-1,j}\lambda_{j-1}(w_{i,j-1}^{n} + w_{i,j-1}^{n+1})$$

$$+ \frac{\Delta t}{2} d_{ij}(n_{ij}^{n} + n_{ij}^{n+1})\} \qquad (3\text{-}14)$$

where, for convenience,

$$\eta_{ij} \equiv e_{ij} + \lambda_j + \sigma_j\phi_i \; . \qquad (3\text{-}15)$$

Applying (3-13) to n_{ij} gives, by means of (3-1),:

$$n_{ij}^{n+1} = n_{ij}^{n} + \frac{\Delta t}{2}\{\frac{dn_{ij}}{dt}|_n + S_{ij}^{n+1} + b_{j-1,j}\lambda_{j-1}n_{i,j-1}^{n+1}$$

$$+ \sum_k \alpha_k f_{kij} n_{kj}^{n+1} + e_{ij}w_{ij}^{n+1} - (\lambda_j+\alpha_i+\sigma_j\phi_i+d_{ij})n_{ij}^{n+1}\}$$

Inserting w_{ij}^{n+1} from (3-14) and rearranging the terms gives:

$$\{1 + \frac{\Delta t}{2}\zeta_{ij} - \left(\frac{\Delta t}{2}\right)^2 \frac{e_{ij}d_{ij}}{1+\frac{\Delta t}{2}\eta_{ij}}\}n_{ij}^{n+1} - \frac{\Delta t}{2}\sum_k \alpha_k f_{kij} n_{kj}^{n+1} \qquad (3\text{-}16)$$

$$= n_{ij}^{n} + \frac{\Delta t}{2}\{\frac{dn_{ij}}{dt}\Big|_n + S_{ij}^{n+1} + b_{j-1,j}\lambda_{j-1}n_{i,j-1}^{n+1} + e_{ij}u_{ij}^{n+1}\}$$

where

$$\zeta_{ij} \equiv \lambda_j + \alpha_i + \sigma_j\phi_i + d_{ij} \qquad (3\text{-}17)$$

and

$$u_{ij}^{n+1} \equiv w_{ij}^{n+1} - \frac{\Delta t}{2} \frac{d_{ij}}{1+\frac{\Delta t}{2}\eta_{ij}} n_{ij}^{n+1}$$

$$= \frac{1}{1+\frac{\Delta t}{2}\eta_{ij}} \{(1-\frac{\Delta t}{2}\eta_{ij})w_{ij}^{n} + \frac{\Delta t}{2} b_{j-1,j}\lambda_{j-1}(w_{i,j-1}^{n}+w_{i,j-1}^{n+1})$$

$$+ \frac{\Delta t}{2} d_{ij} n_{ij}^{n}\} . \qquad (3\text{-}18)$$

Multiplying (3-16) by $\frac{2}{\Delta t}$ gives:

$$(\frac{2}{\Delta t} + \xi_{ij} - \frac{\Delta t}{2} \frac{e_{ij}d_{ij}}{1+\frac{\Delta t}{2}\eta_{ij}}) n_{ij}^{n+1} - \sum_{k} \alpha_k f_{kij} n_{kj}^{n+1}$$

$$= \frac{2}{\Delta t} n_{ij}^{n} + \frac{dn_{ij}}{dt}\Big|_{n} + S_{ij}^{n+1} + b_{j-1,j}\lambda_{j-1} n_{i,j-1}^{n+1} + e_{ij} u_{ij}^{n+1} . \qquad (3\text{-}19)$$

Equations (3-19), and (3-14) also, represent a system of NI x NJ equations. The two systems are liked together in the same manner as the differential equations (3-1) and (3-11), thus forming a system of 2 x NI x NJ equations in the variables n_{ij}^{n+1} and w_{ij}^{n+1}. As such a system easily becomes very large, a special procedure must be applied by the solution to avoid problems with computer storage and time. Figure 3.2 shows schematically the coefficient matrix of the combined system for NJ = 4. Each square symbolizes a submatrix with NI x NI elements. Shading of the squares indicates that the submatrix contains non-zero elements, and dotted or solid lines in the diagonal of some submatrices indicate that these submatrices only have non-zero elements in the diagonal. Empty squares contain noughts only. The equations are arranged with equations (3-19) first and thereafter equations (3-14). Within each of these groups they are arranged in subgroups of constant j-value. By mixing the two sets of equations, an arrangement as illustrated in fig. 3.3 is obtained. Here equations with the same j-value are grouped together. The solution of the equations now proceeds as follows: first the system of NI equations for $n_{i,1}^{n+1}$ is solved; now the terms containing $n_{i,1}^{n+1}$ in the equations for $w_{i,1}^{n+1}$ can be moved to the righthand side, thus

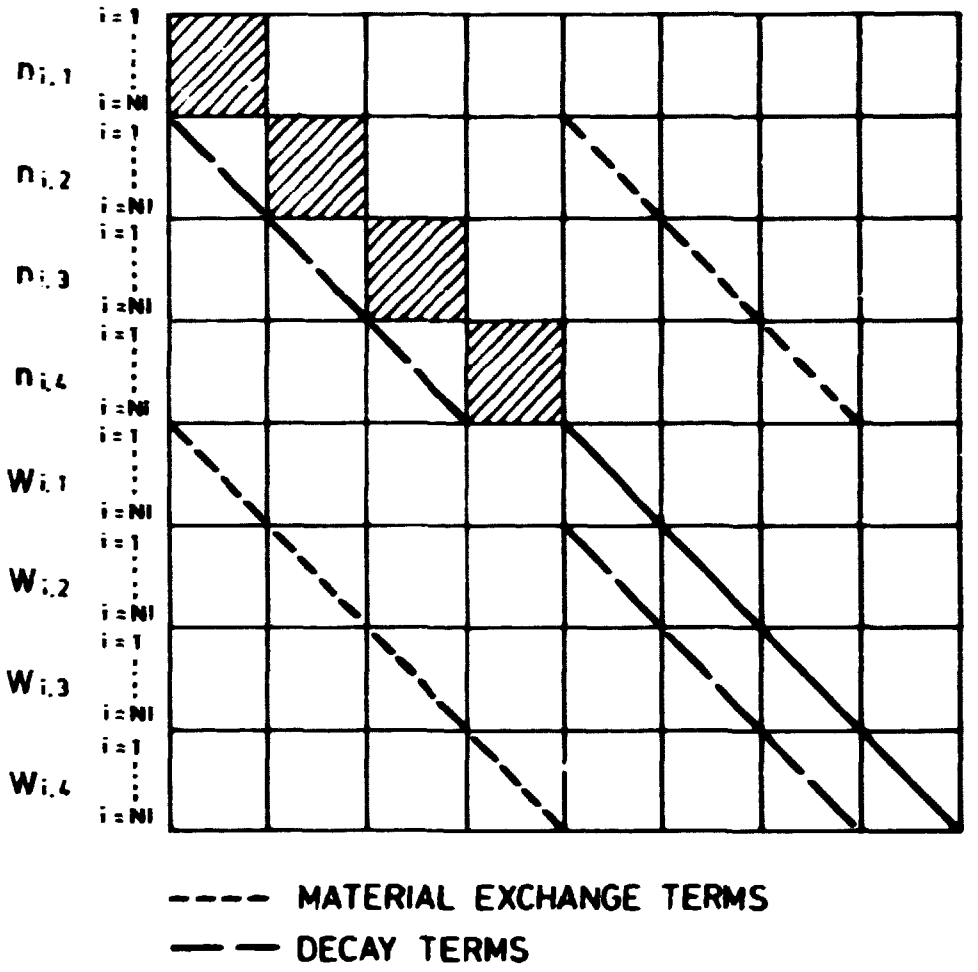

Fig. 3.2. Coefficient matrix.

making possible the straight-forward solution of these equations as already indicated by the expression (3-14). The equations for $n_{i,2}^{n+1}$ are now reduced in the same manner to NI equations with NI variables, which can be solved, and so forth. Using the above-mentioned simple procedure the solution of a large system of 2 x NJ x NI equations becomes the much simpler task of solving 2 x NJ systems each consisting of NI equations. Furthermore, half of these systems - the ones for $w_{i,j}^{n+1}$ - each consist of NI independent equations; physically, this is due to the fact that no coupling exists between the deposits on the surfaces of different components.

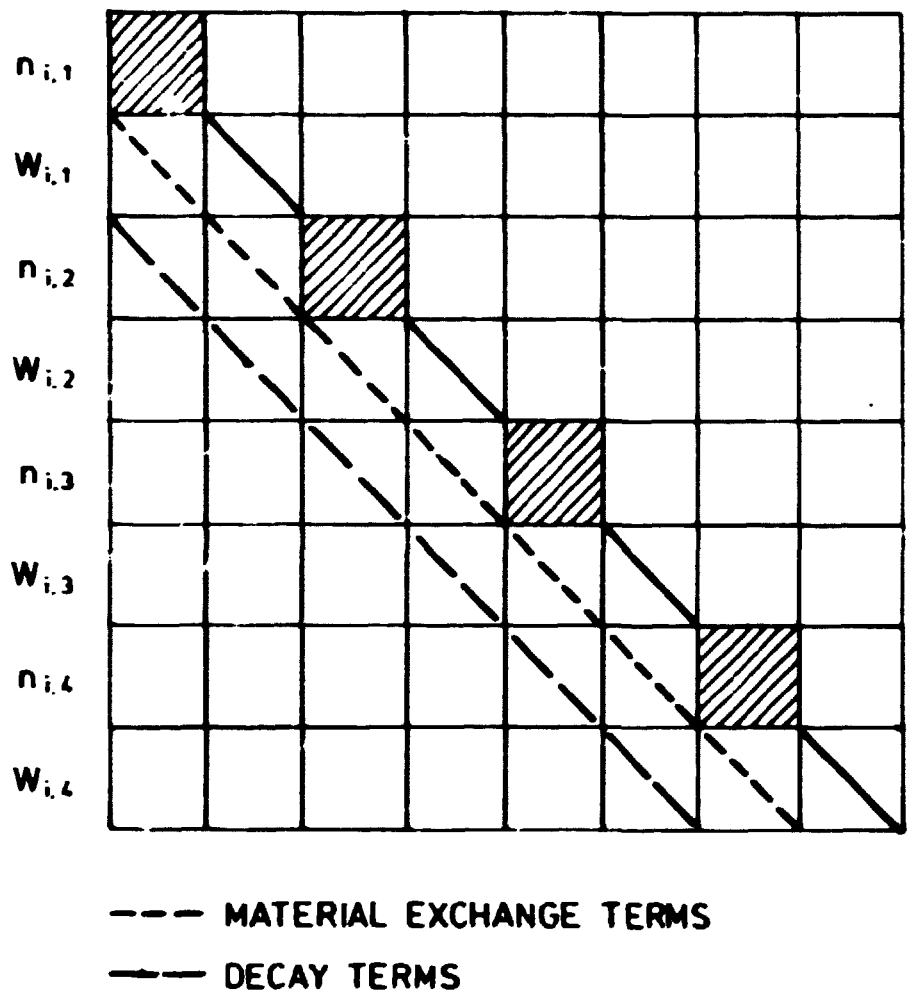

Fig. 3.3. Modified coefficient matrix.

Solving all equations (3-14) and (3-19) yields the values of $w_{i,j}$ and $n_{i,j}$ at time-step n + 1 from the values at step n; thus the integration of the system of differential equations represented by (3-1) and (3-11) consists of NS successive solutions of the 2 x NJ systems of NI equations, where NS denotes the total number of time-steps. NS varies from case to case depending on numerical-error estimates made during the integration, but a typical value of NS would be 150.

3.1.3. Asymptotic Solution

In many cases it is sufficient to know the asymptotic (or steady state) values of n_{ij} and w_{ij}. Equations for these are derived from eqs. (3-1) and (3-11) by setting the derivative equal to zero:

$$\frac{dn_{ij}}{dt} = 0 \quad <=> \quad ,$$

$$S^{\infty}_{ij} + b_{j-1,j}\lambda_{j-1}n^{\infty}_{i,j-1} + \sum_k \alpha_k f_{kij} n^{\infty}_{kj} + e_{ij}w^{\infty}_{ij}$$

$$- (\lambda_j + \alpha_i + \sigma_j\phi_i + d_{ij})\, n^{\infty}_{ij} = 0 \qquad (3\text{-}20)$$

Upper index ∞ denotes asymptotic values.

$$\frac{dw_{ij}}{dt} = 0 \quad <=>$$

$$d_{ij}n^{\infty}_{ij} + b_{j-1,j}\lambda_{j-1}w^{\infty}_{i,j-1} - (e_{ij} + \lambda_j + \sigma_j\phi_i)w^{\infty}_{ij} = 0 \quad <=>$$

$$w^{\infty}_{ij} = \frac{1}{\eta_{ij}} \{d_{ij}n^{\infty}_{ij} + b_{j-1,j}\lambda_{j-1}w^{\infty}_{i,j-1}\} \qquad (3\text{-}21)$$

where η_{ij} is defined by (3-15). Inserting this in (3-20) and using the definition of ξ_{ij} (3-17) gives:

$$S^{\infty}_{ij} + b_{j-1,j}\lambda_{j-1}n^{\infty}_{i,j-1} + \sum_k \alpha_k f_{kij} n^{\infty}_{kj} + \frac{e_{ij}}{\eta_{ij}} b_{j-1,j}\lambda_{j-1}w^{\infty}_{i,j-1}$$

$$+ \left(\frac{1}{\eta_{ij}} e_{ij}d_{ij} - \xi_{ij}\right) n^{\infty}_{ij} = 0$$

Rearranging:

$$\left(\frac{1}{\eta_{ij}} e_{ij}d_{ij} - \xi_{ij}\right)n^{\infty}_{ij} + \sum_k \alpha_k f_{kij} n^{\infty}_{kj}$$

$$= - S^{\infty}_{ij} - b_{j-1,j}\lambda_{j-1} \left(n^{\infty}_{i,j-1} + \frac{e_{ij}}{\eta_{ij}} w^{\infty}_{i,j-1}\right). \qquad (3\text{-}22)$$

Equations (3-21) and (3-22) can be solved by means of the

same procedure as (3-14) and (3-19), yielding the asymptotic values n^{∞}_{ij} and w^{∞}_{ij}.

3.1.4. Conversion Into Units of Radioactivity

Integration of differential equations (3-1) and (3-11), and solution of equations (3-21) and (3-22) yield values of n_{ij} and w_{ij} in terms of number of atoms. These are converted into disintegrations/second by multiplying by λ_j:

$$N_{ij} = \lambda_j n_{ij} \text{ dis/s} \tag{3-23}$$

$$W_{ij} = \lambda_j w_{ij} \text{ dis/s} \tag{3-24}$$

Sometimes it is convenient to express the activity in Curies; this is obtained by dividing by 3.7×10^{10}.

3.2. Shutdowns

When the reactor is shut down, changes in the radioactivity inventories of the various components may occur in several ways. In order to describe the shutdowns in a relatively simple mathematical model, which can give analytical expressions for the activities, a number of assumptions are made concerning the mechanisms involved in a shutdown.

3.2.1. Assumptions

1) Release rate from fuel remains constant.
2) Mass transport stops in turbine system.
3) Total mixing of activity in reactor water system.
4) No transition period exists between operating and shutdown mode.
5) No exchange of material with walls.

A more detailed discussion of these assumptions is given below.

3.2.1.1. Constant Release Rate. It is assumed that the release of fission products from the fuel remains constant throughout the shutdown period - or that the shutdown period can be split up into a number of periods with constant release rate. As will be seen from the equations set up in section

3.2.2., this assumption is a condition for having general analytical expressions for $n_{ij}(t)$ and $w_{ij}(t)$ during shutdown.

3.2.1.2. Stop of Mass Transport in Turbine System. After the reactor has been shut down and steam production has ceased, there will be no significant mass transport in the turbine system components. This assumption removes the links between different components in the mathematical description, thus making it possible to consider each component separately.

3.2.1.3. Total Mixing in Reactor Water. Although steam production stops, some circulation still goes on in the reactor vessel and recirculation system in order to clean the water by passing it through the Reactor Water Clean-Up (RWCU) system. After some time, the inventories of the components containing reactor water will be mixed so that the activity concentrations become equal, except for nuclides with half-lives shorter than the circulation time. However, these nuclides will decay shortly after shutdown and thus be of no importance. The assumption of equal activity concentration leads to the definition of a new "component": Reactor Water, comprising the activity in the water in the following components, cfr. fig. 2.1: Reactor Core, Lower Plenum, Upper Plenum and Separators, RWCU-system, Downcomer, and Recirculation Loops. The removal of activity from the reactor water by the RWCU system is described by means of a "clean-up - constant", cf. the mathematical description in section 3.2.2.

3.2.1.4. No Transition Period. Shutting down a power plant involves a sequence of actions, which may vary from time to time and from plant to plant. The main actions are: stopping the fission process by inserting control rods, bypassing the turbine at an appropriate point of time, providing for residual heat removal, and gradually reducing the coolant flow. A general mathematical description of this transition period is not considered possible. It is therefore anticipated that the duration of the shutdown considered is long enough to allow the neglect of the possible effect of the transition period, unless this can be represented by a simple change in the release rate from the fuel.

Operational experience shows that substantial changes in release rates may occur after shutdown. Measurements from the

Gundremmingen power plant[7] have shown a rise in release rate shortly after shutdown followed by a decrease to about the same level as during operation; from this level a slower decrease was experienced. In 3 out of 4 cases investigated, the peak value occurred 5 hours after shutdown and for ^{131}I it was about a factor of 100 larger than the value during operation. Three to 4 days after shutdown the release rate was about the same as before shutdown.

A detailed description of the transition period has been waived through the assumption of there being no such period. However, the above-mentioned changes in release rate may be accounted for by periods of constant release, as mentioned in subsection 3.2.1.1.

3.2.1.5. No Exchange With Walls. It is assumed that the erosion from and deposition on component surfaces ceases during shutdown. This assumption is obviously reasonable for the erosion term, as this must diminish when the coolant flow decreases or stops. The validity in the case of deposition is more doubtful; for instance, some precipitation of material may occur in stagnant or slowly flowing water. However, the assumption is retained until a better understanding of these phenomena has been achieved.

3.2.2. Mathematical Description

Due to the assumption of no mass transport in the turbine system, each component here can be considered as isolated from the others in the mathematical description. In the reactor water part of the plant the removal of activity by the RWCU system can also be treated without linking any components together. Only when the accumulation of activity in the RWCU system itself is to be calculated, does it become necessary to consider more than one component at a time. However, the transfer of material in this case is a one-way transfer, i.e. there is no feed-back term from the RWCU system. This much simplifies the solution, as will be shown below.

The mathematical description is divided into two steps. First, equations are set up for the release from fuel, decay, and transfer to the RWCU system; later, in subsection 3.2.2.2., various operations such as fuel change and decontamination are treated.

3.2.2.1. Release, Decay, and Clean-up. For components in the turbine system, the number of atoms of nuclide j in the coolant in component i follows the equation

$$\frac{dn_{ij}(t)}{dt} = \lambda_{j-1} b_{j-1,j} n_{i,j-1}(t) - \lambda_j n_{ij}(t). \quad (3\text{-}25)$$

Correspondingly, for deposits on the walls of component i:

$$\frac{dw_{ij}(t)}{dt} = \lambda_{j-1} b_{j-1,j} w_{i,j-1}(t) - \lambda_j w_{ij}(t). \quad (3\text{-}26)$$

These equations are inhomogeneous, first-order linear differential equations having the analytical solutions:

$$n_{ij}(t) = e^{-\int \lambda_j dt} \{ \int \lambda_{j-1} b_{j-1,j} n_{i,j-1}(t) e^{\int \lambda_j dt} dt + C^n_{ij} \} \quad (3\text{-}27)$$

and

$$w_{ij}(t) = e^{-\int \lambda_j dt} \{ \int \lambda_{j-1} b_{j-1,j} w_{i,j-1}(t) e^{\int \lambda_j dt} dt + C^w_{ij} \} \quad (3\text{-}28)$$

where C^n_{ij} and C^w_{ij} are arbitrary constants, and t is the time from shutdown. As shown in detail in Appendix A, $n_{ij}(t)$ and $w_{ij}(t)$ can be expressed as:

$$n_{ij}(t) = \sum_{k=1}^{j} K_{kj} C^n_{ik} e^{-\lambda_k t} \quad (3\text{-}29)$$

and

$$w_{ij}(t) = \sum_{k=1}^{j} K_{kj} C^w_{ik} e^{-\lambda_k t} \quad (3\text{-}30)$$

where

$$\left.\begin{aligned} K_{kj} &\equiv \prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \quad \text{for } k \leq j-1 \\ K_{jj} &\equiv 1 \end{aligned}\right\} \quad (3\text{-}31)$$

$$\left.\begin{aligned} C^n_{ij} &= n_{ij}(0) - \sum_{k=1}^{j-1} K_{kj} C^n_{ik} \quad \text{for } j>1 \\ C^n_{ij} &= n_{ij}(0) \quad \text{for } j=1 \end{aligned}\right\} \tag{3-32}$$

$$\left.\begin{aligned} C^w_{ij} &= w_{ij}(0) - \sum_{k=1}^{j-1} K_{kj} C^w_{ik} \quad \text{for } j>1 \\ C^w_{ij} &= w_{ij}(0) \quad \text{for } j=1 \end{aligned}\right\} \tag{3-33}$$

For the reactor water, the number of atoms for nuclide j is

$$n_{wj}(t) = \sum n_{ij}(t), \tag{3-34}$$

where the sum goes over the components containing reactor water, that is nos. 1, 34, 35, 36, 37, and 38. Due to the assumption of equal mixing, the inventory of one of these components can be expressed as

$$n_{ij}(t) = n_{wj}(t) \cdot \frac{m_i}{m_w}, \tag{3-35}$$

where m_i is the mass of water in component i, and m_w is the total mass of reactor water in the components under consideration; $m_w = \Sigma m_i$. The change in inventory of nuclide j in the reactor water is described by:

$$\frac{dn_{wj}(t)}{dt} = S_j + \lambda_{j-1} b_{j-1,j} n_{w,j-1}(t) - (\lambda_j + \varepsilon_j) n_{wj}(t) \tag{3-36}$$

where ε_j is the clean-up constant determining the rate of the transfer of material to the RWCU system; for convenience, we define $\beta_j \equiv \lambda_j + \varepsilon_j$. Except for the RWCU system, which is treated below, deposits on the walls of components containing reactor water follow the same equations as for deposits in the turbine system. This is due to the assumption of no exchange of material between walls and coolant during shutdown. With constant release from fuel, S_j, eq. (3-36) has the following analytical solution,

which is derived in Appendix A:

$$n_{wj}(t) = \frac{1}{\beta_j} \sum_{k=1}^{j} P_{kj} S_k + \sum_{k=1}^{j} L_{kj} C_{wk}^n \, e^{-\beta_k t} \tag{3-37}$$

where

$$\left.\begin{aligned} P_{kj} &\equiv \prod_{p=k}^{j-1} \frac{\lambda_p}{\beta_p} b_{p,p+1} \qquad \text{for } k \leq j-1 \\ P_{jj} &\equiv 1 \end{aligned}\right\} \tag{3-38}$$

$$\left.\begin{aligned} L_{kj} &\equiv \prod_{p=k}^{j-1} \frac{b_{p,p+1} \lambda_p}{\beta_{p+1} - \beta_k} \qquad \text{for } k \leq j-1 \\ L_{jj} &\equiv 1 \end{aligned}\right\} \tag{3-39}$$

$$C_{wj}^n = n_{wj}(0) - \frac{1}{\beta_j} \sum_{k=1}^{j} P_{kj} S_k - \sum_{k=1}^{j-1} L_{kj} C_{wk}^n \tag{3-40}$$

Material transferred to the RWCU system from the reactor water is deposited on the "walls" of this system, i.e. on filter resins, and is supposed to stay there until the filter resin is changed. The number of atoms of nuclide j on the walls in the RWCU system follows the equation

$$\frac{dw_{Rj}(t)}{dt} = \varepsilon_j n_{wj}(t) + \lambda_{j-1} b_{j-1,j} w_{R,j-1}(t) - \lambda_j w_{Rj}(t) \tag{3-41}$$

Index R denotes the RWCU system. The analytical solution of this equation is a bit more complicated than for the equations considered earlier in this section because of the term $\varepsilon_j n_{wj}(t)$. But, as shown in Appendix A, it is possible to get an analytical solution although the expression becomes somewhat "heavy":

$$w_{Rj}(t) = \frac{1}{\lambda_j} \sum_{m=1}^{j} B_{mj} \frac{\varepsilon_m}{\beta_m} \sum_{k=1}^{m} P_{km} S_k + \sum_{m=1}^{j} \varepsilon_m \Lambda_{mj} \sum_{k=1}^{m} H_{kmj} L_{km} C_{wk}^n e^{-\beta_k t}$$

$$+ \sum_{k=1}^{j} K_{kj} C_{Rk} e^{-\lambda_k t} \tag{3-42}$$

where

$$B_{mj} \equiv \prod_{p=m}^{j-1} b_{p,p+1} \quad \text{for } m \leq j-1;\ B_{jj} \equiv 1 \tag{3-43}$$

$$\Lambda_{mj} \equiv \prod_{p=m}^{j-1} \lambda_p b_{p,p+1} \quad \text{for } m \leq j-1;\ \Lambda_{jj} \equiv 1 \tag{3-44}$$

$$H_{kmj} \equiv \prod_{p=m}^{j} \frac{1}{\lambda_p - \beta_k} \quad \text{for } p \geq k \text{ (always true here)} \tag{3-45}$$

and the other constants are as defined earlier. A sum with an upper bound less than the lower bound is defined as being equal to zero.

$$C_{Rj} = w_{Rj}(0) - \frac{1}{\lambda_j} \sum_{m=1}^{j} B_{mj} \frac{\varepsilon_m}{\beta_m} \sum_{k=1}^{m} P_{km} S_k - \sum_{m=1}^{j} \varepsilon_m \Lambda_{mj} \sum_{k=1}^{m} H_{kmj} L_{km} C_{wk}^{n} - \sum_{k=1}^{j-1} K_{kj} C_{Rk} \tag{3-46}$$

If, for some reason, the reactor water is not cleaned, $\varepsilon_j = 0$ for all nuclides, and the RWCU system is treated like the turbine system components, eq. (3-30).

The differential equations used here for inventories during shutdowns are, in principle, just simplified forms of equations (3-1) and (3-11) and might, as such, have been solved numerically in the same way. However, the analytical approach has been chosen, because it facilitates a more detailed study of the variations in activity with time. In fact, if one just knows the inventories at the start of the shutdown period, the inventories at any time during the shutdown are easily evaluated. If the assumption of constant S_j should prove too coarse, an analytical solution would still be possible, if S_j can be expressed as a "simple" function of time - i.e. a function that does not make the evaluation of the integrals containing S_j impossible. If this is not possible, or if non-negligible mass transport continues during the shutdown, the analytical description must be abandoned. Equations (3-1) and (3-11) will then be used to describe the behaviour during shutdown, changing parameters such as inventory change rate and partition factors.

Figure 3.4 illustrates the usefulness of being able to evaluate the activity at any time during shutdown. The rise in ^{135}Xe -activity would not be discovered if one just calculated the activity before and after shutdown.

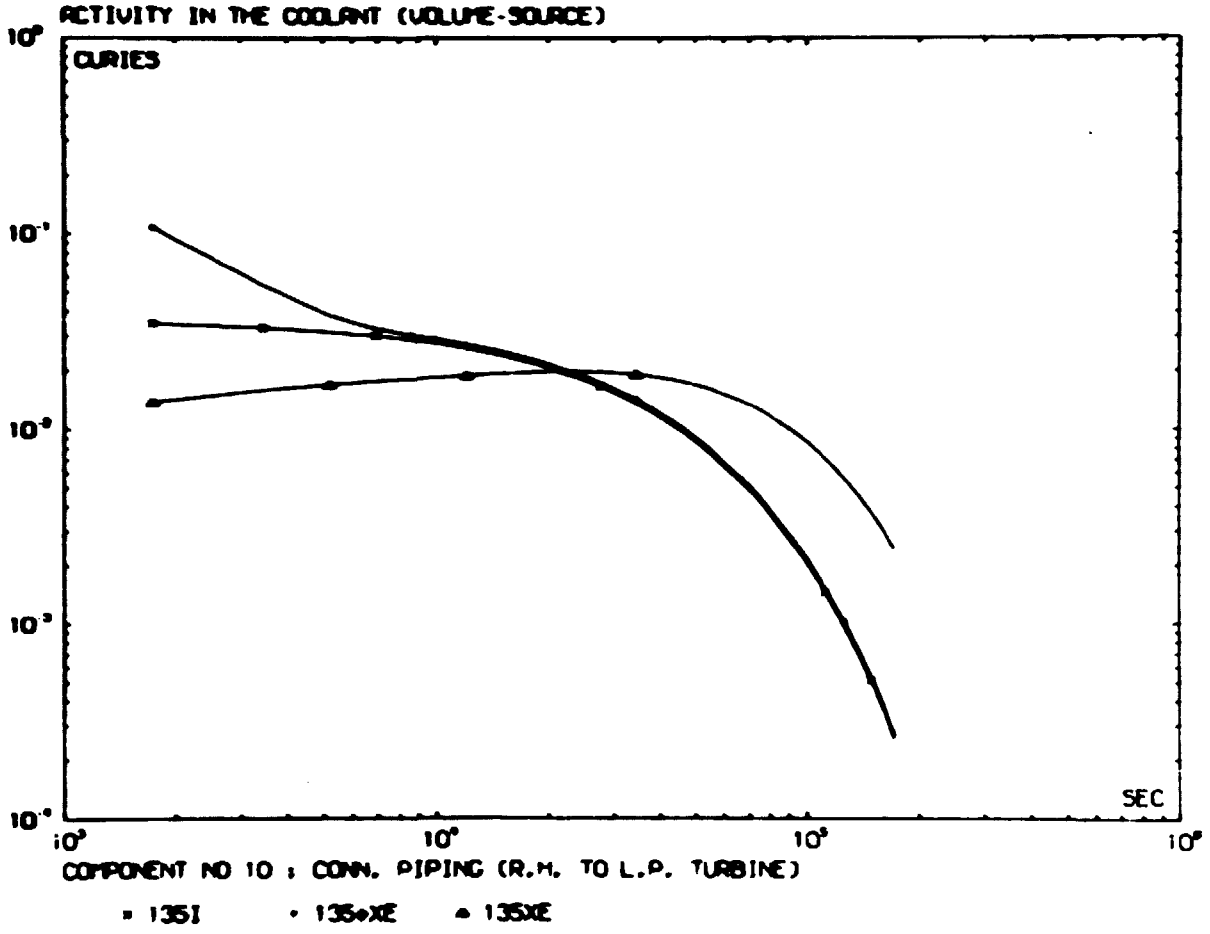

Fig. 3.4. Activity during shutdown.

3.2.2.2. Changes and Decontamination. During a shutdown period various maintenance and repair jobs may be performed, including the decontamination of some components and replacement of others by new, clean ones. Furthermore, fuel change is performed once a year. These measures are accounted for in the following manner. If a fraction, FR, of a component is replaced, the activity in deposits on the walls of this component is reduced by the same fraction:

$$w_{after} = (1-FR) \cdot w_{before}. \qquad (3-47)$$

The decay term evaluated in the previous section is superposed upon this change. It is supposed that the component has to be emptied when a change occurs, so that the activity in the coolant is reduced to zero for components undergoing changes. Decontamination is treated in the same way as the replacement of a fraction of a component. The "change fraction", FR, is easily evaluated from the decontamination factor DF:

$$\left.\begin{aligned} DF &\equiv \frac{w_{before}}{w_{after}} \\ w_{after} &= (1-FR) \cdot w_{before} \end{aligned}\right\} \Rightarrow$$

$$\frac{1}{1-FR} = DF \Rightarrow FR = 1 - \frac{1}{DF}$$

When fuel is changed, the activity in the reactor water is not supposed to be affected in other ways than those described in the previous section, i.e. decay, change in fission product release, and maybe an increased clean-up rate.

4. MODEL FOR CORROSION PRODUCTS

Corrosion of reactor materials and the subsequent release, transport, and deposition of corrosion products are of interest from several viewpoints in relation to nuclear power plants. First of all, the mechanical integrity of plant components should not be affected by corrosion. Neither is it tolerable that deposits on fuel rods grow too thick, as this increases the thermal resistance of the cladding and decreases the flow area, thereby increasing the risk of burn-out. Finally, corrosion products may become activated and in the form of so-called crud move around suspended in the coolant or settle on component surfaces.

Due to the above-mentioned circumstances, much work has been devoted to investigating the mechanisms behind corrosion and the release/deposition of corrosion products, and to collecting data

concerning the extent of these phenomena in existing nuclear power plants. Experiments have been performed in order to determine the corrosion rates of various reactor materials under different conditions, changing parameters such as pH, and temperature. Generally measurements have been made on so-called coupons - small pieces of the material to be examined that are placed in the desired environment for a period of time and thereafter removed for examination. This method yields an average of the corrosion rate during the test period, but does not give much information about the corrosion rate as a function of time. However, until recently these results were the only ones available[8-10], Newer experiments[11,12] have used on-line counting on activated corrosion products such as ^{60}Co, ^{51}Cr, and ^{59}Fe, thereby giving a time-dependent picture of the build-up of corrosion products. This seems to indicate that corrosion of steels follows either parabolic or logarithmic functions of time, i.e. expressions of the types

$$m(t) = (K_p t + m_o^2)^{\frac{1}{2}} \qquad (4\text{-}1)$$

and

$$m(t) = K_\ell \ln (a_\ell t + 1) \qquad (4\text{-}2)$$

where $m(t)$ is the amount of oxide per unit area (gm^{-2}) at time t, and K_p, K_ℓ, a_ℓ, and m_o are constants that are determined by fitting data from measurements to the above expressions. The logarithmic expression (4-2) seems to give the best fit to data for steel[12]. From (4-1) and (4-2) the corrosion rate at any time is easily evaluated:

$$\frac{dm(t)}{dt} = \frac{1}{2} K_p (K_p t + m_o^2)^{-\frac{1}{2}} \qquad (4\text{-}3)$$

or

$$\frac{dm(t)}{dt} = \frac{K_\ell a_\ell}{a_\ell t + 1} \; . \qquad (4\text{-}4)$$

Models concerning the deposition of corrosion products on component surfaces and the release from these deposits have been proposed by a number of authors[5,6,13-15]. Bartlett[15] gives a

thorough theoretical discussion of the fundamental processes that are assumed to govern the deposition and release, and develops a system of equations describing these processes. However, these equations contain a number of parameters that are not very well determined - or not known at all. So until this situation improves, simpler models[5,6,13,14] will still prevail. These models have in common that they describe the exchange of material between bulk coolant and component walls by means of first-order coefficients, which are derived from experiments or from measurements on reactors. The model used in the present work utilizes such first-order coefficients - as was the case for fission products in the preceding chapter.

The mathematical model to be set up for corrosion product inventories posesses great similarity to that for fission products. Indeed, it will be shown that the equations can be solved by the same computer code. As was the case for fission products, the description is here divided into two sections: 1) normal operation, and 2) shutdowns.

4.1. Normal Operation

The fission product equations have to describe decay-chains consisting of a number of nuclides ranging, typically, from 1 to 7. Equations for corrosion products only have to describe two nuclides at a time: a non-radioactive target nuclide and its activation product nuclide, e.g. ^{59}Co and ^{60}Co. Theoretically, the activation product nuclide might capture a neutron, too, but this process is not considered to be of importance here and consequently no "secondary" activation products are taken into account. For each component in the plant model (cf. fig. 2.1), the number of corrosion product atoms in the coolant is described by the equations:

$$\frac{dn_{i1}}{dt} = Q_{i1}(t) + \sum_k \alpha_k f_{ki1} n_{k1} + e_i w_{i1} - (\alpha_i + \sigma_1 \phi_i + d_i) n_{i1} \tag{4-5}$$

$$\frac{dn_{i2}}{dt} = Q_{i2}(t) + \sigma_1 \phi_i n_{i1} + \sum_k \alpha_k f_{ki2} n_{k2} + e_i w_{i2} - (\lambda_2 + \sigma_2 \phi_i + \alpha_i + d_i) n_{i2}. \tag{4-6}$$

The number of corrosion product atoms deposited on component surfaces is described by the equations:

$$\frac{dw_{i1}}{dt} = d_i n_{i1} - (\sigma_1 \phi_i + e_i) w_{i1} \qquad (4\text{-}7)$$

$$\frac{dw_{i2}}{dt} = d_i n_{i2} + \sigma_1 \phi_i w_{iI} - (\lambda_2 + \sigma_2 \phi_i + e_i) w_{i2} \qquad (4\text{-}8)$$

where index 1 denotes the non-radioactive target nuclide and index 2 the activated nuclide. $Q_{i1}(t)$ and $Q_{i2}(t)$ are source terms, which will be discussed below. As is seen, the above equations are essentially the same as those for fission products (3-1) and (3-11). Thus many of the terms have already been discussed and will only be mentioned briefly in the following discussion.

4.1.1. Discussion of the Equations

4.1.1.1. Source Term. The terms $Q_{i1}(t)$ and $Q_{i2}(t)$ represent the emission of corrosion products from the construction material of component number i into the coolant. They are not to be confused with the terms $e_i w_{ij}$, which represent the release of material deposited in a film on the surfaces of component i. $Q_{i1}(t)$ is the emission rate of non-radioactive corrosion product nuclides, which are then carried with the coolant and undergo activation when passing through the reactor core. $Q_{i2}(t)$ is the emission rate of nuclides that have been activated while still in the construction material; this term will only assume non-zero values for components with a neutron flux, i.e. the reactor core and perhaps the downcomer. The emission rate of non-radioactive nuclides in component i is expressed as

$$Q_{i1}(t) = \frac{N_{Av}}{M_{A,s}} a_j^s \sum_m A_{m,i} R_{m,i}(t) p_m^s \qquad \text{(atoms/s)} \qquad (4\text{-}9)$$

where

N_{Av} = the Avogadro number (mol^{-1}),

$M_{A,s}$ = the atomic mass of element s (kg/mol),

a_j^s = the isotopic abundance of isotope j (the one under consideration) of element s (fraction of atoms),

$A_{m,i}$ = the surface area of construction material m in component i (m^2),

$R_{m,i}(t)$ = the emission rate for corrosion products of material m in component i ($kg\ m^{-2}s^{-1}$),

p_m^s = the weight fraction of element s in construction material m, and

the sum goes over all construction materials used in component i.

The emission rate $R_{m,i}(t)$ will have the same time dependence as the corrosion rate, assuming that a fixed fraction of the corroding material is released at any point of time. So whenever data are available, the logarithmic corrosion kinetics represented by eq. (4-4) will be used. Otherwise, a constant value of the emission rate will be applied.

During reactor operation, radioactive nuclides are created in the construction material of the core and the downcomer. Let n_1 be the number of target nuclides supposed to remain constant. The number of nuclides activated from these, n_2, follows the equation

$$\frac{dn_2(t)}{dt} = \sigma_1 \phi n_1 - \lambda_2 n_2(t) \qquad (4\text{-}10)$$

with the solution

$$n_2(t) = \frac{1}{\lambda_2} \sigma_1 \phi n_1 + Ce^{-\lambda_2 t} \qquad (4\text{-}11)$$

where

$$C = (n_2(t_1) - \frac{1}{\lambda_2} \sigma_1 \phi n_1) e^{\lambda_2 t_1}, \qquad (4\text{-}12)$$

and t_1 is the time at which the reactor starts operation.

During shutdown periods the activation ceases, and the number of active nuclides follows the equation

$$\frac{dn_2(t)}{dt} = -\lambda_2 n_2(t) \Rightarrow n_2(t) = n_2(t_o) e^{-\lambda_2 (t-t_o)} \qquad (4\text{-}13)$$

where t_o denotes the time for shutdown.

It is assumed that the emission rate does not depend on whether the oxides contain the target nuclides or their acti-

vation product nuclides. So during reactor operation the emission rate of activated nuclides from the construction material in components with neutron flux is expressed as

$$Q_{i2}(t) = Q_{i1}(t) \cdot \frac{n_2(t)}{n_1}$$

$$= Q_{i1}(t) \cdot \{\frac{1}{\lambda_2}\sigma_1\phi + (\frac{n_2(t_1)}{n_1} - \frac{1}{\lambda_2}\sigma_1\phi)e^{-\lambda_2(t-t_1)}\}$$

$$= Q_{i1}(t)\{\frac{1}{\lambda_2}\sigma_1\phi + (\frac{Q_{i2}(t_1)}{Q_{i1}(t_1)} - \frac{1}{\lambda_2}\sigma_1\phi)e^{-\lambda_2(t-t_1)}\} \quad (4\text{-}14)$$

The emission rate of activated nuclides during a shutdown starting at time t_o is

$$Q_{i2}(t) = Q_{i1}(t)\frac{n_2(t)}{n_1} = Q_{i1}(t)\frac{n_2(t_o)}{n_1}e^{-\lambda_2(t-t_o)}$$

$$= Q_{i1}(t)\{\frac{1}{\lambda_2}\sigma_1\phi + (\frac{Q_{i2}(t_1)}{Q_{i1}(t_1)} - \frac{1}{\lambda_2}\sigma_1\phi)e^{-\lambda_2(t_o-t_1)}\}e^{-\lambda_2(t-t_o)}$$

$$\equiv Q_{i1}(t)\, F_i(t_o,t_1)e^{-\lambda_2(t-t_o)}. \quad (4\text{-}15)$$

Table 4.1 summarizes some properties of five of the most common construction materials, and table 4.2 gives important parameters for those activation products that have proved to be of greatest significance in operating reactors. As will be noticed from table 4.2, most of the activated corrosion products have rather long halflives. This means that they will stay radioactive for a long period after reactor shutdown, thus constituting a radiation hazard to maintenance and repair crews. As most fission products are relatively shortlived, corrosion products will dominate after some days of shutdown.

4.1.1.2. Decay Terms. Contrary to eqs. (3-1) and (3-11) for fission products, eqs. (4-5) to (4-8) do not contain terms describing the production of some nuclides by the decay of others. Here the radioactive nuclide is produced by neutron absorption in the target nuclide, described by the terms $\sigma_1\phi_1 n_{i1}$ and $\sigma_1\phi_1 w_{i1}$. This process was also introduced as a possibility for

Table 4.1

Some properties of frequently occurring construction materials

Material	Composition (weight %)	Applications (typical)	Corrosion rate (10^{-11} kg $m^{-2}s^{-1}$)	Release rate (10^{-11} kg $m^{-2}s^{-1}$	Surface area in BWR/6 pressure vessel(m^2)
304 Stainless Steel	Ni:8-11, Cr:18-20 Mn:2.0 max, C:0.08 max, S:0.03, Si:0.75 max, P:0.04 max, Fe:balance	Inside cladding of pressure vessel, recirculation system components; control rods	10-19 $\frac{4.09 \cdot 10^3}{4.56 \cdot 10^{-5}t+1}$ *)	2-6	∿ 3200
Carbon steel	C:0.08, Mn:0.3 P:0.01, S:0.02 Si:<0.02, Cr:0.012 Ni:0.008, Cu:0.03, Fe:balance	Pressure vessel, steam piping	40-100	∿ 40	∿ 60
Zircaloy-2	Sn:1.2-1.7,Fe:0.07-0.2,Ni:0.03-0.08,Cr: 0.05-0.15, Zr:balance	Fuel element cladding	4-30	very low	7527
Zircaloy-4	Sn:1.2-1.7, Fe:0.18-0.24, Ni:0.007 max, Cr: 0.07-0.13, Zr:balance	Fuel channel	4-30	very low	3415
Inconel-X	C:0.04, Mn:0.5, Si:0.4, Cr:15.0, Ni:73.0, Nb: 1.0, Ti:2.5, Fe:7.0, Al:0.7	Springs for fuel rod spacers	4-10	4-6	223

*) eq. (4-4). Constants are from [12]

Table 4.2

Important radioactive corrosion products

Parent nuclide	Abundance of parent (%)	Nuclear reaction	Cross section (barns)	Product nuclide	Half-life	Gamma energy (MeV)	Abundance of gamma energies (fraction)
^{50}Cr	4.4	n,γ	16.0 ± 0.5^{x}	^{51}Cr	27.8 d	0.32	0.09
^{54}Fe	5.9	n,p	$(82.5\pm5)\cdot10^{3}$ *	^{54}Mn	303 d	0.84	1.0
^{55}Mn	100	n,γ	13.3 ± 0.1^{x}	^{56}Mn	2.57 h	0.85 1.81 2.11	0.99 0.23 0.14
^{58}Ni	68.0	n,p	$(113\pm7)\cdot10^{-3}$ *	^{58}Co	71.3 d	0.81	0.99
^{59}Co	100	n,γ	$37,5 \pm 0.2^{x}$	^{60}Co	5.2 y	1.17 1.33	1.0 1.0
^{60}Ni	26.2	n,p	$(2.3\pm0.4)\cdot10^{-3}$*				
^{58}Fe	0.33	n,γ	1.14 ± 0.05 x	^{59}Fe	45 d	0.19 1.1 1.29	0.03 0.56 0.44
^{63}Cu	69.1	n,γ	4.4 ± 0.2 x	^{64}Cu	12.8 h	1.34	0.01
^{64}Zn	48.9	n,γ	0.82 ± 0.01 x	^{65}Zn	245 d	1.12	0.49

* Fission spectrum averaged, taken from [19].

x Thermal (2200 m/s), taken from [19].

fission products, and in practice it is treated by the computer code in the same way as decay. The terms $-\lambda_2 n_{i2}$ and $-\lambda_2 w_{i2}$ describe the removal of activation product nuclides by decay.

4.1.1.3. Mass Transport Terms. Mass transport is described in the same way for corrosion products as in the case of fission products by the terms $-\alpha_i n_{ij}$ and $\sum_k \alpha_k f_{kij} n_{kj}$.

4.1.1.4. Exchange With Walls. The build-up of deposits on component surfaces and the release of material from these deposits is described by the terms $d_i n_{ij}$ and $e_i w_{ij}$ as was the case for fission products, too. However, the evaluation of the activity in deposits is considered to be slightly more reliable for corrosion products, as these have been used for most measurements concerning deposition/release. Nevertheless, it must be admitted that this area is one with great differences between measured data. This is illustrated by table 4.3 that gives values of the deposition and release constants as reported by different authors.

Table 4.3

Deposition and release constants

Nuclide used in experiment	d (s^{-1})	e (s^{-1})	Reference
^{51}Cr	$1.3 \cdot 10^{-4}$	$4 \cdot 10^{-9}$	6
^{51}Cr	$8.2 \cdot 10^{-5}$	$8.2 \cdot 10^{-12}$	16
^{59}Fe	$6 \cdot 10^{-2}$	$2.6 \cdot 10^{-7}$	6
^{59}Fe	$1 \cdot 10^{-3}$	$1.9 \cdot 10^{-8}$	16
^{59}Fe	-	$2.8 \cdot 10^{-6}$	5
^{60}Co	$7 \cdot 10^{-2}$	$2.3 \cdot 10^{-7}$	6
^{60}Co *	$9 \cdot 10^{-3}$	$2.6 \cdot 10^{-7}$	6
^{60}Co	$2.5 \cdot 10^{-4}$	-	16
^{64}Cu	$9.4 \cdot 10^{-6}$	$2.8 \cdot 10^{-6}$	6
^{95}Zr	$2.6 \cdot 10^{-3}$	$2.4 \cdot 10^{-11}$	16

*From measurements on zircaloy surfaces. The other data are measured on surfaces of various types of steel.

4.1.1.5. Neutron Absoption Terms. The generation of radioactive corrosion product nuclides takes place by neutron absorption as already mentioned in subsection 4.1.1.2. Furthermore, some of the active nuclides may be removed by neutron absorption, represented by the terms $-\sigma_2\Phi_i n_{i2}$ and $-\sigma_2\Phi_i w_{i2}$ in eqs. (4-6) and (4-8). These terms also describe the creation of a new nuclide which may be radioactive, but, as mentioned previously, it is assumed that nuclides created by this "secondary activation" can be neglected. The product of cross-section and neutron flux used in the absorption terms, of course, represents the integrated value over all energies:

$$\sigma\Phi \equiv \int_0^\infty \sigma(E)\Phi(E)dE \qquad (4\text{-}16)$$

where $\Phi(E)$ is the neutron flux per unit energy interval.

4.1.2. Integration of the Equations

Comparison shows that eqs. (4-5) and (4-6) are of the same form as eq. (3-1), and that eqs. (4-7) and (4-8) are of the same form as eq. (3-11). Accordingly, the procedures described in section 3.1.2. for the integration and section 3.1.3. for the asymptotic solution will apply to the corrosion product equations as well.

4.1.3. Conversion of Units

The solutions of the differential equations will be n_{i1}, n_{i2}, w_{i1}, and w_{i2} given in terms of number of atoms. The amount of the activation product nuclide, denoted by index 2, should be expressed in dis/s or Ci:

$$N_{i2} = n_{i2} \cdot \lambda_2 \text{ dis/s} = n_{i2} \cdot \lambda_2 \cdot \frac{1}{3{,}7\cdot 10^{10}} \text{ Ci} \qquad (4\text{-}17)$$

$$W_{i2} = w_{i2} \cdot \lambda_2 \text{ dis/s} = w_{i2} \cdot \lambda_2 \cdot \frac{1}{3{,}7\cdot 10^{10}} \text{ Ci} \qquad (4\text{-}18)$$

Related to radiation field calculations, the amounts of inactive nuclides are, of course, of little interest, but they emerge as a by-product in this evaluation. The masses of the element s are

$$N_{is} = n_{il} \cdot \frac{M_{As}}{N_{Av} a_j^s} \quad kg \tag{4-19}$$

$$W_{is} = w_{il} \cdot \frac{M_{As}}{N_{Av} a_j^s} \quad kg \tag{4-20}$$

where M_{As} and a_j^s are as defined in subsection 4.1.1.1.

4.2. Shutdowns

As was the case for fission products, a simple mathematical model with an analytical solution will be set up for corrosion products in order to describe the inventory changes in detail as a function of time. A number of assumptions, almost identical to those for fission products, are set up:

4.2.1. Assumptions

1) The emission rate remains constant or follows a simple function of time.
2) Mass transport stops in the turbine system.
3) Total mixing of material suspended in the reactor water.
4) No transition period between operating and shutdown mode.
5) No exchange of material with walls.

The discussion of most of these points has been given in section 3.2.1. and will not be repeated here, only a few supplementary comments are needed.

The emission rate appears to be even more unpredictable during a shutdown than during normal operation. This is due to the fact that changes occur in many of the parameters influencing - in a more or less well known way - the corrosion and emission rates. For instance, large increases in the emission rate - so-called crud bursts - occur due to rapid changes in temperature, flow rate, pH, and oxygen content[17,18]. At the least, temperature and flow rate are changed during every shutdown, but not nescessarily at the same rate every time. Thus crud bursts may occur during some shutdowns and not in others.

In this model it is assumed that the emission rate can be described as a simple function of time, i.e. one that does not prevent the evaluation of the integral set up in the following section. The simplest case, of course, appears when the emission rate keeps constant during part of or the entire shutdown.

4.2.2. Mathematical Description

As was the case for fission products, the mathematical description is divided into two parts, the first part treating emission, decay, and clean-up, and the second part dealing with decontamination, fuel change, etc.

4.2.2.1. Emission, Decay, and Clean-up. For the components in the turbine system, the number of atoms of the non-radioactive target nuclide in the coolant will be determined by:

$$\frac{dn_{il}(t)}{dt} = Q_{il}(t) \quad \Rightarrow \quad n_{il}(t) = n_{il}(t_o) + \int_{t_o}^{t} Q_{il}(\tau)d\tau \qquad (4\text{-}21)$$

where t_o denotes the time of shutdown.

If the emission rate $Q_{il}(t)$ is constant during the shutdown, the solution is simple:

$$n_{il}(t) = n_{il}(t_o) + (t - t_o) \cdot Q_{il}(t_o), \qquad (4\text{-}22)$$

and if, for instance, the emission follows logarithmic kinetics, the solution is (cf. equations (4-2), (4-4), and (4-9)):

$$n_{il}(t) = n_{il}(t_o) + \int_{t_o}^{t} \frac{N_{Av}}{M_{As}} a_j^s \sum_m A_{m,i} p_m^s \frac{K_{\ell,m}\, a_{\ell,m}}{a_{\ell,m}\tau + 1} d\tau$$

$$= n_{il}(t_o) + \frac{N_{Av}}{M_{As}} a_j^s \sum_m A_{m,i} p_m^s K_{\ell,m} \ln\left(\frac{a_{\ell,m}t + 1}{a_{\ell,m}t_o + 1}\right). \qquad (4\text{-}23)$$

Due to the assumption of no exchange with walls,

$$\frac{dw_{il}(t)}{dt} = 0 \Rightarrow w_{il}(t) = w_{il}(t_o). \qquad (4\text{-}24)$$

For the activated nuclide, the emission rate is zero in turbine system components as there is no activation here. So the number of atoms in the coolant and on the walls will be determined by

$$\frac{dn_{i2}(t)}{dt} = -\lambda_2 n_{i2}(t) \quad \Rightarrow \quad n_{i2}(t) = n_{i2}(t_o)e^{-\lambda_2(t-t_o)} \tag{4-25}$$

$$\frac{dw_{i2}(t)}{dt} = -\lambda_2 w_{i2}(t) \quad \Rightarrow \quad w_{i2}(t) = w_{i2}(t_o)e^{-\lambda_2(t-t_o)} \tag{4-26}$$

The inventories of the components containing reactor water are assumed to be mixed like the fission product inventories. Thus the number of atoms in the reactor water is

$$n_{w1}(t) = \sum_{RWC} n_{i1}(t) \text{ and } n_{w2}(t) = \sum_{RWC} n_{i2}(t) \tag{4-27}$$

where the sums go over the "Reactor Water Components", i.e. component nos. 1, 34, 35, 36, 37, and 38. The inventory of one of these components is

$$n_{i1}(t) = n_{w1}(t)\frac{m_i}{m_w} \text{ and } n_{i2}(t) = n_{w2}(t)\frac{m_i}{m_w} \tag{4-28}$$

where m_i is the mass of reactor water in component i and m_w is the total mass of reactor water, $m_w = \sum_{RWC} m_i$. The number of atoms of the inactive nuclide in the reactor water follows the equation

$$\frac{dn_{w1}(t)}{dt} = Q_{w1}(t) - \varepsilon_1 n_{w1}(t) \quad \Rightarrow$$

$$n_{w1}(t) = e^{-\varepsilon_1 t}\{\int Q_{w1}(t)e^{\varepsilon_1 t}\,dt + C_1\}, \tag{4-29}$$

where $Q_{w1}(t)$ is the total emission rate of nuclide 1, the inactive nuclide, from the construction materials in the reactor water system,

$$Q_{w1}(t) = \sum_{RWC} Q_{i1}(t). \tag{4-30}$$

The clean-up rate for nuclide no. 1 is ε_1. Equation (4-29) is only valid for $\varepsilon_1 > 0$. If $\varepsilon_1 = 0$ the solution becomes

$$n_{w1}(t) = n_{w1}(t_o) + \int_{t_o}^{t} Q_{w1}(\tau)d\tau \qquad (4\text{-}31)$$

If the emission rates $Q_{i1}(t)$ keep constant during the shutdown, the solutions become simple:

$$n_{w1}(t) = \frac{1}{\varepsilon_1} Q_{w1}(t_o) + (n_{w1}(t_o) - \frac{1}{\varepsilon_1} Q_{w1}(t_o))e^{-\varepsilon_1(t-t_o)} \qquad (4\text{-}32)$$

and for $\varepsilon_1 = 0$:

$$n_{w1}(t) = n_{w1}(t_o) + (t - t_o)\, Q_{w1}(t_o). \qquad (4\text{-}33)$$

If the emission follows logarithmic kinetics, it is still possible but a bit time-consuming to evaluate an expression for $n_{w1}(t)$. Due to this fact, and because only constant emission rates have been applied in the computer code so far, the evaluation is not given here but in Appendix B.

The amount of the target nuclide deposited on the walls of components in the reactor water system is assumed not to change during shutdown, so

$$\frac{dw_{w1}(t)}{dt} = 0 \Rightarrow w_{w1}(t) = w_{w1}(t_o). \qquad (4\text{-}34)$$

For the activation product nuclide, the following equation applies in the reactor water system

$$\begin{aligned}\frac{dn_{w2}(t)}{dt} &= \sum_{PWC} Q_{i2}(t) - \beta_2 n_{w2}(t) \\ &= \sum_{RWC} Q_{i1}(t)F_i(t_o,t_1)e^{-\lambda_2(t-t_o)} - \beta_2 n_{w2}(t) \qquad (4\text{-}35)\end{aligned}$$

where $\beta_2 \equiv \lambda_2 + \varepsilon_2$, and ε_2 is the clean-up rate for the activation product nuclide. Usually $\varepsilon_2 = \varepsilon_1$. $F_i(t_o,t_1)$ is defined by eq. (4-15). If the emission rate for inactive nuclides keeps constant, (4-35) can be integrated. For convenience, we first define

$$Q_{w2}(t_o) \equiv \sum_{RWC} Q_{i1}(t_o)F_i(t_o,t_1).$$

$$n_{w2}(t) = e^{-\beta_2 t}\{\int Q_{w2}(t_o)e^{-\lambda_2(t-t_o)}e^{\beta_2 t}dt + C_2\} \qquad (4\text{-}36)$$

$$= e^{-\beta_2 t}Q_{w2}(t_o)e^{\lambda_2 t_o}\int e^{\varepsilon_2 t}dt + C_2e^{-\beta_2 t}.$$

For $\varepsilon_2 = 0$:

$$n_{w2}(t) = e^{-\lambda_2 t}Q_{w2}(t_o)e^{\lambda_2 t_o}t + C_2e^{-\lambda_2 t}$$

$$= e^{-\lambda_2(t-t_o)}Q_{w2}(t_o)t + \{(n_{w2}(t_o)$$

$$- Q_{w2}(t_o)t_o)e^{\lambda_2 t_o}\}e^{-\lambda_2 t}$$

$$= n_{w2}(t_o)e^{-\lambda_2(t-t_o)} + (t-t_o)Q_{w2}(t_o)e^{-\lambda_2(t-t_o)}. \qquad (4\text{-}37)$$

For $\varepsilon_2 > 0$:

$$n_{w2}(t) = \frac{1}{\varepsilon_2}Q_{w2}(t_o)e^{\lambda_2 t_o}e^{-\beta_2 t}e^{\varepsilon_2 t} + C_2e^{-\beta_2 t}$$

$$= \frac{1}{\varepsilon_2}Q_{w2}(t_o)e^{-\lambda_2(t-t_o)} + C_2e^{-\beta_2 t}$$

$$= \frac{1}{\varepsilon_2}Q_{w2}(t_o)e^{-\lambda_2(t-t_o)} + \{(n_{w2}(t_o)$$

$$- \frac{1}{\varepsilon_2}Q_{w2}(t_o))e^{\beta_2 t_o}\}e^{-\beta_2 t}$$

$$= n_{w2}(t_o)e^{-\beta_2(t-t_o)} + \frac{1}{\varepsilon_2} Q_{w2}(t_o)\{e^{-\lambda_2(t-t_o)} - e^{-\beta_2(t-t_o)}\}. \quad (4\text{-}38)$$

The integration of eq. (3-35) in the case of logarithmic emission kinetics is treated in Appendix B. The deposits on the walls of components in the reactor water system - excluding the RWCU system which is treated below - follow the equation

$$\frac{dw_{w2}(t)}{dt} = -\lambda_2 w_{w2}(t) \quad => \quad (4\text{-}39)$$

$$w_{w2}(t) = w_{w2}(t_o)e^{-\lambda_2(t-t_o)}. \quad (4\text{-}40)$$

If there is no cleaning of the reactor water, $\varepsilon_1 = \varepsilon_2 = 0$. In this case the deposits on the "walls", i.e. the filters, of the RWCU system will follow equations (4-34) and (4-40). When cleaning does take place, the deposits for non-active nuclides follow the equation

$$\frac{dw_{R1}(t)}{dt} = \varepsilon_1 n_{w1}(t) \quad => \quad (4\text{-}41)$$

$$w_{R1}(t) = w_{R1}(t_o) + \int_{t_o}^{t} \varepsilon_1 n_{w1}(\tau)\, d\tau \quad (4\text{-}42)$$

When the emission rates $Q_{i1}(t)$ are constant during shutdown, $n_{w1}(t)$ is expressed by (4-32), and $w_{R1}(t)$ can be evaluated:

$$w_{R1}(t) = w_{R1}(t_o) + \int_{t_o}^{t} \varepsilon_1 \{\frac{1}{\varepsilon_1} Q_{w1}(t_o) + (n_{w1}(t_o) - \frac{1}{\varepsilon_1} Q_{w1}(t_o))e^{-\varepsilon_1(\tau-t_o)}\} d\tau$$

$$= w_{R1}(t_o) + (t-t_o)Q_{w1}(t_o) - (n_{w1}(t_o) - \frac{1}{\varepsilon_1} Q_{w1}(t_o))(e^{-\varepsilon_1(t-t_o)} - 1) \quad (4\text{-}43)$$

The activation product nuclides deposited in the RWCU system during shutdown follow the equation

$$\frac{dw_{R2}(t)}{dt} = \varepsilon_2 n_{w2}(t) - \lambda_2 w_{R2}(t) \quad \Rightarrow \tag{4-44}$$

$$w_{R2}(t) = e^{-\lambda_2 t}\{\varepsilon_2 \int n_{w2}(t) e^{\lambda_2 t} dt + C_R\} \tag{4-45}$$

If the emission rates $Q_{i1}(t)$ stay constant during shutdown, (4-45) becomes, cf. (4-38):

$$w_{R2}(t) = e^{-\lambda_2 t} \varepsilon_2 \int \{n_{w2}(t_o) e^{-\beta_2 (t-t_o)}$$

$$+ \frac{1}{\varepsilon_2} Q_{w2}(t_o)(e^{-\lambda_2 (t-t_o)} - e^{-\beta_2 (t-t_o)})\} e^{\lambda_2 t} dt + C_R e^{-\lambda_2 t}$$

$$= e^{-\lambda_2 t} \varepsilon_2 n_{w2}(t_o) e^{\beta_2 t_o} \int e^{-\varepsilon_2 t} dt + e^{-\lambda_2 t} Q_{w2}(t_o) e^{\lambda_2 t_o} \int dt$$

$$- e^{-\lambda_2 t} Q_{w2}(t_o) e^{\beta_2 t_o} \int e^{-\varepsilon_2 t} dt + C_R e^{-\lambda_2 t}$$

$$= -n_{w2}(t_o) e^{-\beta_2 (t-t_o)} + e^{-\lambda_2 (t-t_o)} Q_{w2}(t_o) \cdot t$$

$$+ e^{-\beta_2 (t-t_o)} \cdot \frac{1}{\varepsilon_2} Q_{w2}(t_o) + C_R e^{-\lambda_2 t}$$

$$= -\{n_{w2}(t_o) - \frac{1}{\varepsilon_2} Q_{w2}(t_o)\} e^{-\beta_2 (t-t_o)} + Q_{w2}(t_o) \cdot t \cdot e^{-\lambda_2 (t-t_o)}$$

$$+ \{w_{R2}(t_o) + n_{w2}(t_o) - \frac{1}{\varepsilon_2} Q_{w2}(t_o) - Q_{w2}(t_o) \cdot t_o\} e^{-\lambda_2 (t-t_o)}$$

$$= w_{R2}(t_o) e^{-\lambda_2 (t-t_o)} + \{n_{w2}(t_o) - \frac{1}{\varepsilon_2} Q_{w2}(t_o)\}\{e^{-\lambda_2 (t-t_o)}$$

$$- e^{-\beta_2 (t-t_o)}\} + Q_{w2}(t_o) \ (t-t_o) e^{-\lambda_2 (t-t_o)} \tag{4-46}$$

The integration of eqs.(4-42) and (4-45) in the case of logarithmic corrosion kinetics is performed in Appendix B.

4.2.2.2. Changes and Decontamination. The procedure for treating decontamination and the replacement of components or parts thereof is the same for corrosion products as that applied for fission products. So the change in activity in the deposits on component walls due to replacements or decontamination is expressed as

$$w_{after} = (1 - FR)w_{before} \qquad (4\text{-}47)$$

where FR is the fraction replaced. In the case of decontamination

$$FR = 1 - \frac{1}{DF} . \qquad (4\text{-}48)$$

The activity of the coolant in components where changes occur is reduced to zero, as it is assumed that these components have to be emptied during the change.

5. INTRINSIC ACTIVATION PRODUCTS

Radioactive nuclides that are produced by activation of the coolant itself are referred to as intrinsic activation products. For light-water-cooled reactors, the most prominent of these products is ^{16}N, which is produced by an (n,p)-reaction with ^{16}O. Due to its short halflife of 7.4s and high gamma-energies (6.1 and 7.1 MeV), this nuclide dominates the radiation field around reactor and turbine equipment during operation. After shutdown it is of no significance. Data for ^{16}N and other intrinsic activation product nuclides are given in table 5.1. Only ^{16}N and ^{18}O are gamma emitters, so during normal operation when the coolant is kept inside the closed coolant circuit only these two nuclides will be of interest. The two long-lived beta emitters ^{3}H and ^{14}C are most important considered as parts of the liquid and gaseous effluents from the plant. For a 1000 MWe BWR, the production rate of tritium due to activation of the reactor water will be about 0.3 Ci/s and the equilibrium content in the reactor water will be about 10 Ci, dependent on flow rate. This source is a minor one compared to the formation of tritium by ternary fission and by the process $^{10}B(n,2\alpha)^{3}H$ in control rods. These two processes yield of the order of 1000 and 700 times more tritium than the activation of deuterium[20]. However, the tritium produced by these processes is not released to the reactor water and thus poses no problem to the operating staff of the plant. The release to the environment will take place from the reprocessing and waste-handling plants. It must be noted that the figures given above apply to boiling water reactors only.

The production rate of ^{14}C from activation of ^{17}O in the reactor water is about 16 Ci/y for a 1000 MWe BWR. About 11 Ci/y is formed by activation of ^{17}O in the UO_2 fuel pellets; this part is released from the reprocessing plant. Due to the very long halflife of this nuclide, the activity released to the atmosphere will accumulate, and on a global basis this could give rise to some concern. For comparison, it can be mentioned that due to cosmic radiation about 30 kCi/y of ^{14}C is formed in the Earth's atmosphere.

In the following section a simple model for calculation of the inventories of intrinsic activation products will be presented.

Table 5.1

Intrinsic activation products

Parent nuclide	Abundance of parent (%)	Nuclear reaction	Cross section (10^{-3} barn)	Threshold energy (MeV)	Product nuclide	Half-life	Gamma energy (MeV)	Abundance of gammas (fraction)
^{2}H	0.01492	(n,γ)	0.5 ± 0.1[x]	-	^{3}H	12.26 y	-	-
^{16}O	99.759	(n,p)	0.019±0.001[•]	10.24	^{16}N	7.4 s	6.13 7.10	0.69 0.05
^{17}O	0.037	(n,p)	0.0086±0.0008[•]	8.36	^{17}N	4.2 s	-	-
^{17}O	0.037	(n,α)	235 ± 10[x]	-	^{14}C	5760 y	-	-
^{18}O	0.204	(n,γ)	0.16 ± 0.01[x]	-	^{19}O	26.8 s	1.6	0.7

[x] Thermal

[•] Fission spectrum averaged, taken from [19]

5.1. Calculation of Inventories

Consider a mass-unit of coolant entering a fuel channel. The activation of nuclides as a function of distance from the bottom of the channel is described by the equation

$$\frac{dn^*(z)}{dz} = \frac{dn^*(z)}{dt}\,\frac{dt}{dz} = \{\sigma\Phi(z)\frac{N_{Av}a_j}{M_M} - \lambda n^*(z)\}\,\frac{1}{v(z)} \qquad (5\text{-}1)$$

where

$n^*(z)$ = the number of activated nuclides per mass unit (atoms/kg), $n^*(0) = 0$,

σ = the absorption cross section of the parent (cm^2)

$\Phi(z)$ = the average neutron flux in the channel considered at the distance z from inlet ($cm^{-2}s^{-1}$),

N_{Av} = the Avogadro number (atoms/mol),

a_j = the isotopic abundance of the parent nuclide (fraction)

M_M = the molecular weight of the coolant (kg/mol),

λ = the decay constant (s^{-1}), and

$v(z)$ = the coolant velocity at the distance z from the inlet (ms^{-1}).

The coolant velocity can be expressed by means of the density $\rho(z)$:

$$v(z) = \frac{\dot{M}_C}{\rho(z)\cdot A_{ch}} \qquad (5\text{-}2)$$

where $\dot{M}_C$ is the mass flow (kg/s) of coolant and A_{ch} is the flow area (m^2) for the fuel channel under consideration. Introducing (5-2) into (5-1) gives

$$\frac{dn^*(z)}{dz} = \sigma\,\frac{N_{Av}a_j}{M_M}\,\frac{A_{ch}}{\dot{M}_C}\,\rho(z)\Phi(z) - \lambda\,\frac{A_{ch}}{\dot{M}_C}\,\rho(z)n^*(z). \qquad (5\text{-}3)$$

This differential equation usually has to be solved numerically, which is easily done provided that the functions $\rho(z)$ and $\Phi(z)$ are given - for instance, as a polynomial expansion or as a table.

The differential equation (5-3) applies to one fuel channel; so, to evaluate the activation in the whole core, the integration should be repeated for all 732 fuel channels. Another method is to use radially averaged values of ρ and ϕ and let (5-3) be the equation for the whole core, replacing A_{ch} and $\dot{M}_c$ by the corresponding values for the whole core, A_c and $\dot{M}$. Generally, the latter method will be preferable and adequate.

Integration of eq. (5-3) from z = 0 (core inlet) to z = H (core outlet) yields the increase in the concentration of radioactive nuclides by passage of the core. At equilibrium, the inlet concentration is determined by the expression

$$n(o) = n(H) \sum_r \frac{\dot{M}_r}{\dot{M}} e^{-\lambda T_r} \qquad (5\text{-}4)$$

where n(z) denotes the total number of active nuclides per unit mass at the distance z from the core inlet. T_r is the time for "one water molecule" to pass from the core outlet, through the system along the flow path denoted by index r, and back to the core inlet. There are two essentially different flow paths, as indicated in figure 5.1, namely through the turbine system and through the recirculation system. Comparison with figure 2.1 reveals that the turbine system represents a number of different paths, but the main part of the coolant entering this system follows the "long" route through the condenser. Furthermore, the recirculation flow will dominate in this context due to the shorter passage time and the larger mass flow rate.

Let n*(H) be the solution of eq. (5-3). Then the concentration at the core outlet is

$$\begin{aligned} n(H) &= n(0)e^{-\lambda T_c} + n^*(H) \\ &= n(H) \sum_r \frac{\dot{M}_r}{\dot{M}} e^{-\lambda(T_r+T_c)} + n^*(H) \\ &= n^*(H) \left\{1 - \sum_r \frac{\dot{M}_r}{\dot{M}} e^{-\lambda(T_r+T_c)}\right\}^{-1} \end{aligned} \qquad (5\text{-}5)$$

where T_c is the core passage time.

If the neutron flux is assumed to be constant throughout the core, eq. (5-1) for the single channel can be replaced by the simpler equation for the whole core

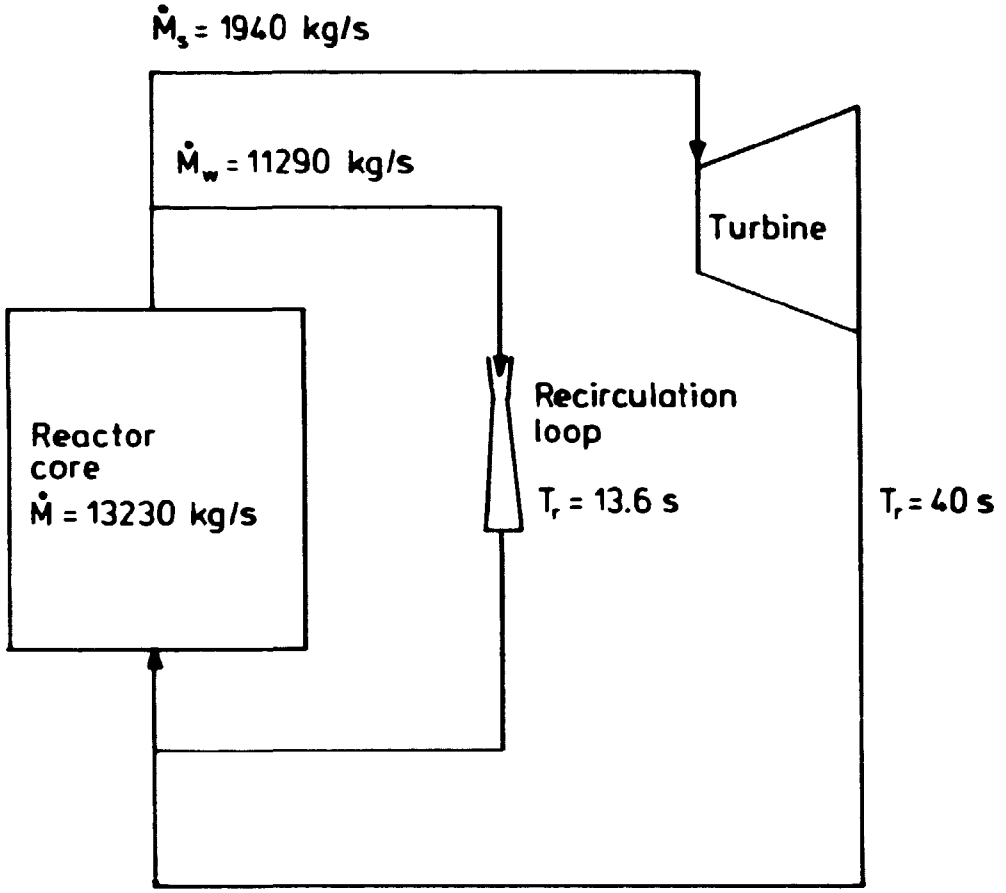

Fig. 5.1. Flow paths for the coolant.

$$\frac{dn(t)}{dt} = \sigma\Phi \frac{N_{Av} a_j}{M_M} - \lambda n(t) \quad , \; o \leq t \leq T_c . \tag{5-6}$$

At equilibrium the solution is

$$n(T_c) = \frac{\sigma\Phi}{\lambda} \frac{N_{Av} a_j}{M_M} \{1-e^{-\lambda T_c}\} \{1-\sum_r \frac{\dot{M}_r}{\dot{M}} e^{-\lambda (T_r+T_c)}\}^{-1} . \tag{5-7}$$

$$n(T_c) \equiv n(H).$$

The concentration of radioactive nuclides as expressed by (5-5) and (5-7) is converted to activity concentration by multiplying with the decay constant:

$$a(z) = \lambda n(z) \text{ dis/kg·s}$$

$$= \frac{\lambda}{3.7 \cdot 10^{10}} n(z) \text{ Ci/kg.} \qquad (5\text{-}8)$$

Often it is convenient to use the flow rate of activity:

$$\dot{a}(z) = a(z) \cdot \dot{M} \text{ Ci/s.} \qquad (5\text{-}9)$$

For a component outside the reactor core, the activity inventory is determined in the following way

$$\frac{dA_i(t)}{dt} = \dot{a}_{in}(t) - \dot{a}_{out}(t) - \lambda A_i(t). \qquad (5\text{-}10)$$

Assuming steady state conditions, we have (cf. fig. 5.2):

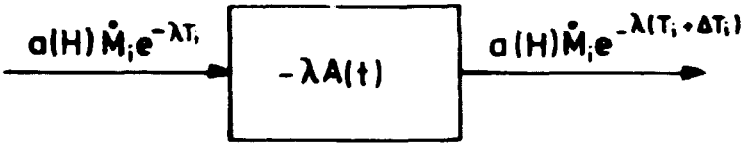

Fig. 5.2. Activity flow in component i.

$$\frac{dA_i}{dt} = 0 = a(H)\dot{M}_i e^{-\lambda T_i} - a(H)\dot{M}_i e^{-\lambda (T_i + \Delta T_i)} - \lambda A_i \Rightarrow$$

$$A_i = \frac{1}{\lambda} a(H)\dot{M}_i e^{-\lambda T_i}\{1 - e^{-\lambda \Delta T_i}\} \qquad (5\text{-}11)$$

where

A_i = the activity in component i at steady state (Ci),

$\dot{M}_i$ = the mass flow rate in component i (kg/s),

T_i = the flow time from core outlet to the inlet of component i (s), and

ΔT_i = the passage time for component i (s).

It has been assumed in eqs. (5-4) and (5-11) that the concentrations of activation product nuclides in water and steam were

equal at any point in the circuit. According to measurements performed on the EBWR[21], this is not always true. At power levels ranging from 5 to 30 MW thermal, the ratio of ^{16}N concentration in steam to ^{16}N concentration in water varied between 0.05 and 1. However, the ratio seemed to grow with the power level, which speaks in favour of assuming equal concentrations in a 3500 MW(th) reactor. Should this assumption not hold, no drastic modifications are needed in the equations. Let a_s and a_w be the activity concentrations in steam and water, respectively, and $\dot{M}_s$ and $\dot{M}_w$ the corresponding mass flow rates at the core outlet. The following correlations then apply to the activity distribution between steam and water

$$\left.\begin{aligned} & a_s\dot{M}_s + a_w\dot{M}_w = \dot{M}a(H) \\ & f_{sw} = \frac{a_s}{a_w} \end{aligned}\right\} \Rightarrow$$

$$a_w = a(H)\frac{\dot{M}}{f_{sw}\dot{M}_s+\dot{M}_w} \equiv a(H)f_w \quad \text{and} \quad a_s = f_{sw}a_w \equiv a(H)f_s \qquad (5\text{-}12)$$

In eq. (5-4) the mass flow rate $\dot{M}_r$ for paths through the turbine system should be multiplied by f_s, and $\dot{M}_r$ for the recirculation system path with f_w. Similarly in eq. (5-11) a_i must be multiplied by f_s for turbine system components and by f_w for reactor water components.

6. SHIELDING CALCULATIONS

In order to assess the radiation fields around components, it is necessary to know how much attenuation of the radiation is provided by the walls of these components and the shielding, if any. It is a difficult task to calculate this attenuation with great accuracy, especially for components with complex geometries, e.g. turbines. In this study only a very simple procedure has been used so far. In the following section a description is given of the method that is based on the point kernel technique applying build-up factors. The source geometry is assumed to be cylindrical, i.e. the equations set up will apply for tubes and other

cylindrical components and for components which are approximated by a cylinder. Only gamma radiation is considered.

6.1. Description of the Method

Figure 6.1 shows a cylindrical volume source of length H and radius R placed in a coordinate system so that the z-axis becomes the cylinder axis and the one end of the cylinder is at z = 0. The point P_B is the observation point, i.e. the point

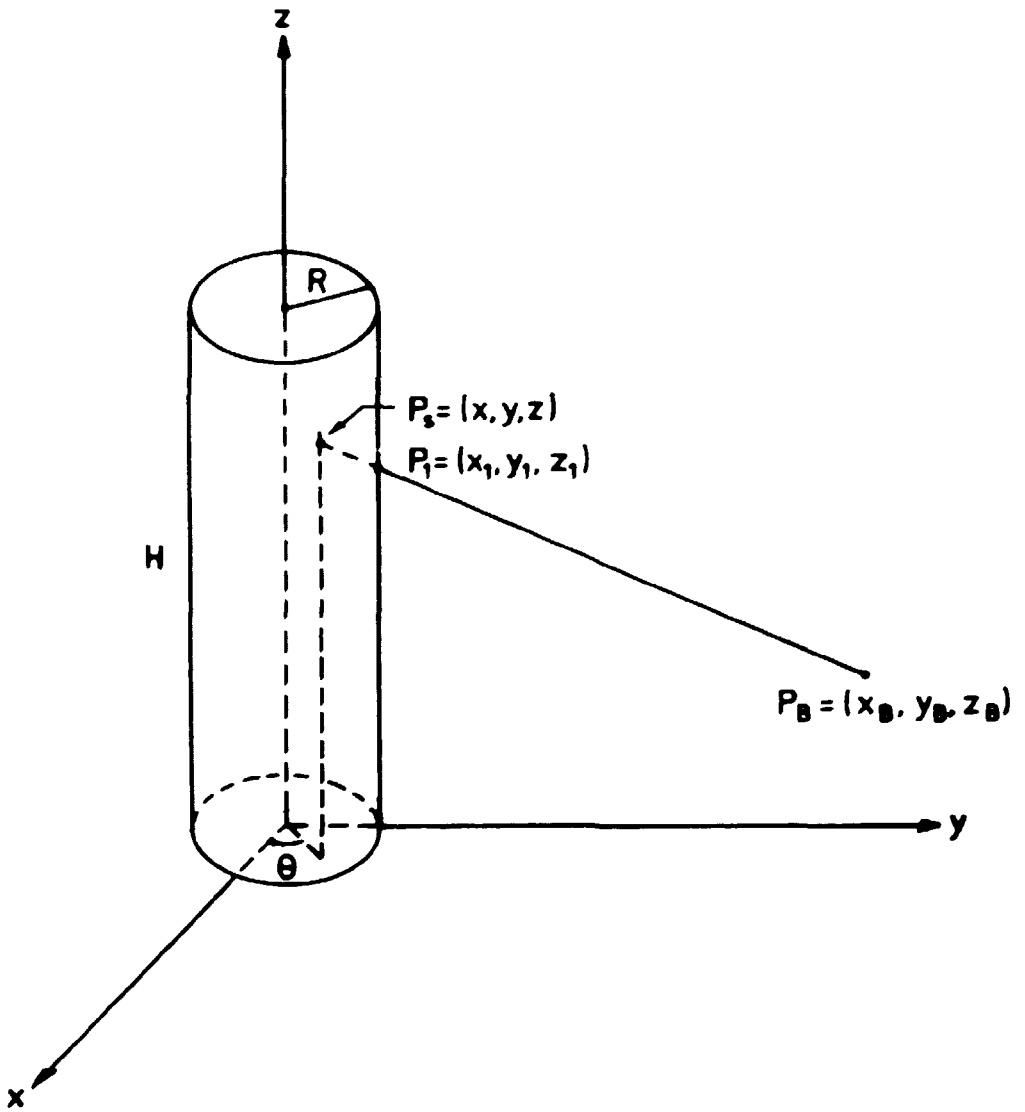

Fig. 6.1. Cylinder source.

where the dose rate is to be calculated. P_S is a source point inside the cylinder and P_1 is the intersection of the cylinder surface and a line from P_S to P_B. The cylinder coordinates of

P_S, which are used in the integration over the source volume, are (r,θ,z) and we have the well known correlations between these and the right-angled coordinates:

$$x = r\cos\theta \text{ and } y = r\sin\theta.$$

The source may be shielded by a number of layers of different material, e.g. the steel tube and an extra concrete shield.

The dose rate in air at point P_B is evaluated by the expression

$$\dot{D}_B = \left(\frac{\mu_E}{\rho}\right)_{air} E\, P_E\, \Phi_B \qquad (6\text{-}1)$$

where

$$\Phi_B = 3.7\cdot10^{10} \int_V S_V(r,\theta,z)\, B(r,\theta,z) \exp\left(-\sum_n \mu_n d_n(r,\theta,z)\right) \cdot \frac{1}{4\pi R_B(r,\theta,z)^2} dV$$

and

μ_E = the energy absorption coefficient for air (cm^{-1}),

ρ = the density of air (g/cm^3),

E = the photon energy (MeV),

P_E = the fraction of decays having the energy E,

V = the source volume (m^3),

$S_V(r,\theta,z)$ = the source strength at point (r,θ,z) (Ci/m^3),

$B(r,\theta,z)$ = the dose build-up factor (see section 6.2),

μ_n = the absorption coefficient of shielding layer no. n (cm^{-1})-the source is counted as layer no. 1,

$d_n(r,\theta,z)$ = the optical distance in layer no. n for the ray from P_S to P_B (cm), and

$R_B(r,\theta,z)$ = the distance from P_S to P_B (cm).

If the source is a surface source, i.e. deposits on the walls, Φ_B is expressed as

$$\phi_B = 3.7 \cdot 10^{10} \int_A S_S(\theta,z) \cdot B(\theta,z) \exp(-\sum_n \mu_n d_n(\theta,z)) \frac{1}{4\pi R_B(\theta,z)^2} dA \qquad (6\text{-}3)$$

where

A = the surface area of the source (m^2), and

$S_S(\theta,z)$ = the source strength at point (R,θ,z) (Ci/m^2).

The integrals in (6-2) and (6-3) have to be evaluated numerically. In order to do this the parameters depending on the source point coordinates (r,θ,z) have to be expressed as functions of these coordinates - or of (x,y,z), which is often more convenient. The source strength is constant in most cases; if not, it is assumed to be specified as a function of (x,y,z) in the computer code. The build-up factor is a function of the optical distance travelled by the gamma ray in the different materials and will be treated in section 6.2. The distance from the source point to the observation point, R_B, is expressed by

$$R_B^2 = (x_B-x)^2 + (y_B-y)^2 + (z_B-z)^2. \qquad (6\text{-}4)$$

The optical distance, d_n, in layer no. n is evaluated below for the three cases: cylindrical shield, slab shield parallel to the cylinder axis, and slab shield perpendicular to the cylinder axis.

6.1.1. Cylindrical Shield

Figure 6.2 shows a projection on the (x,y)-plane of a cylinder source with two layers of shielding. The points P_n' are the projections of the intersection points P_n between the outer surface of layer n and a line connecting the source point and the observation point. Correspondingly, d_n' is the projection of the distance d_n travelled in layer n by the "direct ray".

The distance is expressed as

$$d_n = \sqrt{(x_n-x_{n-1})^2 + (y_n-y_{n-1})^2 + (z_n-z_{n-1})^2} \quad \text{for } n > 1 \qquad (6\text{-}5)$$

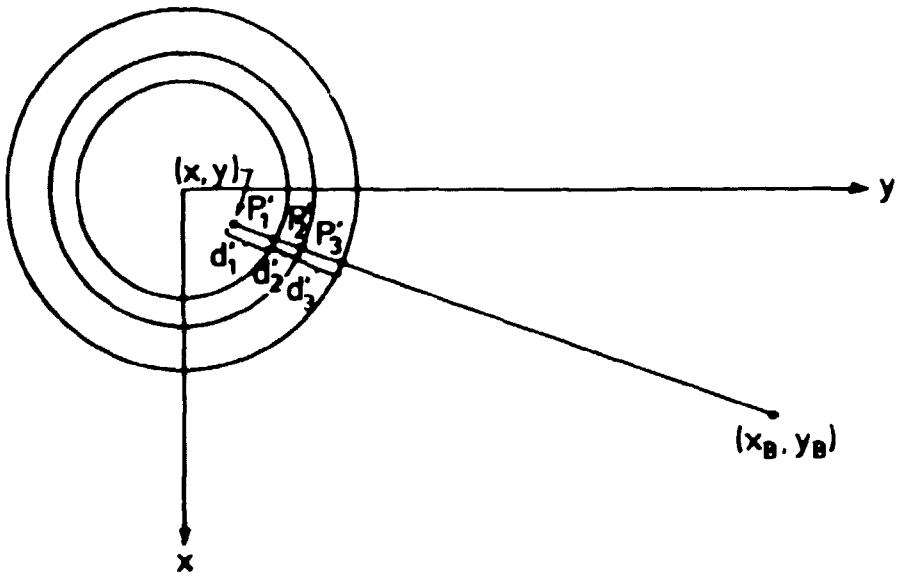

Fig. 6.2. Projection on (x,y)-plane. Cylindrical shields.

and

$$d_1 = \sqrt{(x_1-x)^2 + (y_1-y)^2 + (z_1-z)^2}. \tag{6-6}$$

The coordinates (x_n,y_n,z_n) are found by means of simple geometry. Let R_n be the outside radius of layer number n ($R_1 \equiv R$). Then

$$x_n^2 + y_n^2 = R_n^2 \tag{6-7}$$

In the following we distinguish between the two cases: $x \neq x_B$ and $x = x_B$.

1) $x \neq x_B$:

$$y_n-y = (y_B-y)\frac{x_n-x}{x_B-x} \equiv K(x_n-x) \tag{6-8}$$

$$z_n-z = (z_B-z)\frac{x_n-x}{x_B-x}\ . \tag{6-9}$$

K is defined by eq. (6-8). Inserting y_n from (6-8) into (6-7):

$$x_n^2 + (y+K(x_n-x))^2 = R_n^2 \Rightarrow$$

$$(1+K^2)x_n^2 + 2K(y-Kx)x_n + (y-Kx)^2 - R_n^2 = 0 \Rightarrow$$

$$x_n = \frac{-K(y-Kx) \pm \sqrt{K^2(y-Kx)^2 + (1+K^2)(R_n^2-(y-Kx)^2)}}{(1+K^2)} \qquad (6\text{-}10)$$

The two solutions for x_n correspond to the two possible intersections of a circle and a line. The one with a + before the square root is called x_{n+}, the other x_{n-}. Using fig. 6.2, and the fact that $x_{n+} > x_{n-}$, it is easily verified that

$x_n = x_{n+}$ for $x < x_B$ and

$x_n = x_{n-}$ for $x > x_B$.

Now that x_n is known, y_n and z_n are found from (6-8) and (6-9).

2) $x = x_B$:

In this case $x_n = x$. Inserting this into (6-6) gives:

$$y_n^2 = R_n^2 - x^2 \Rightarrow$$

$$y_n = \pm\sqrt{R_n^2-x^2}.$$

By means of fig. 6.2 it is easily verified that y_n and y_B must have the same sign when $x_n = x = x_B$. Thus

$$y_n = \sqrt{R_n^2-x^2} \quad \text{for } y_B > 0 \qquad (6\text{-}11)$$

$$y_n = -\sqrt{R_n^2-x^2} \quad \text{for } y_B < 0. \qquad (6\text{-}12)$$

Now z_n can be found from the relation

$$z_n - z = (z_B - z)\frac{y_n - y}{y_B - y} . \qquad (6\text{-}13)$$

The special case $x = x_B \wedge y = y_B$ is only of academic interest. It occurs when both the source point and the observation point are placed at the inside wall of the first shielding layer.

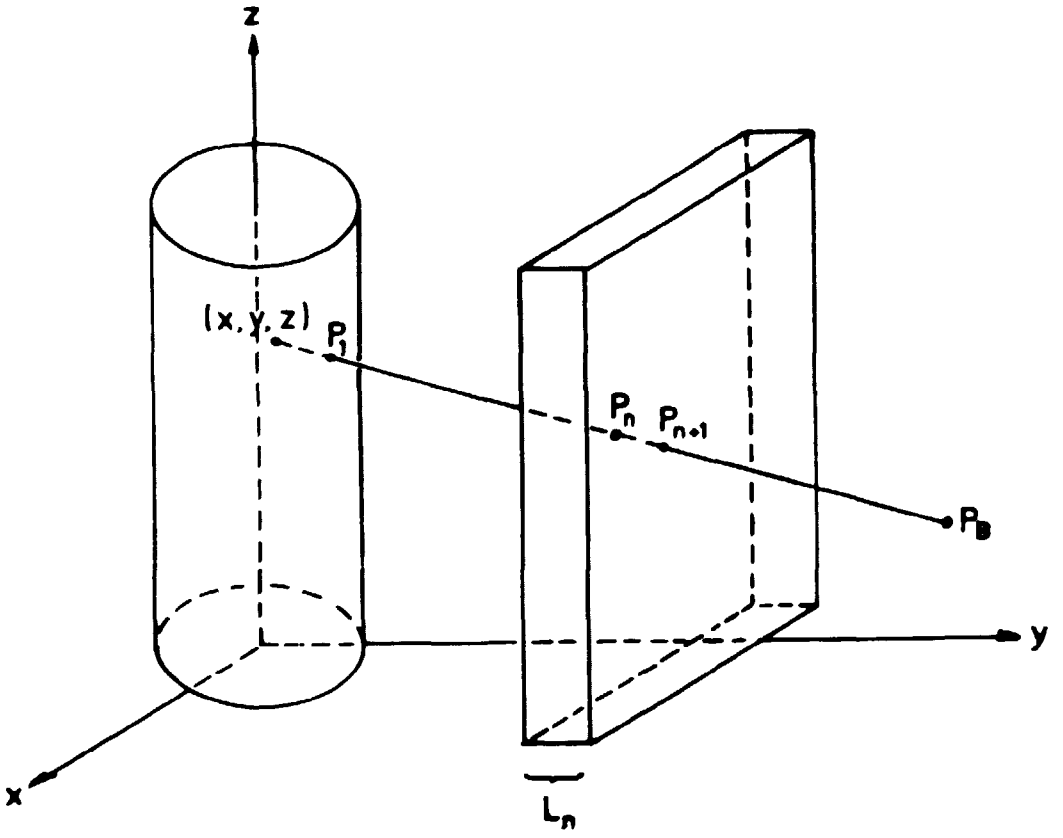

Fig. 6.3. Slab shield parallel to the cylinder axis.

6.1.2. Slab Shield Parallel to the Cylinder Axis

Figure 6.3 shows a slab shield of thickness L_n being penetrated by a ray from the source point (x,y,z) to the observation point $P_B = (x_B,y_B,z_B)$. The distance travelled in the slab is

$$d_n = \sqrt{(x_{n+1}-x_n)^2 + (y_{n+1}-y_n)^2 + (z_{n+1}-z_n)^2} \qquad (6\text{-}14)$$

For convenience, it will be assumed that the coordinate system is arranged so that the x-axis is perpendicular to the slab. The only restriction introduced hereby is that different slabs have

to be parallel, if there is more than one slab shield. Now d_n can be expressed in a simpler way:

$$d_n = L_n \frac{R_B}{x_B - x} \tag{6-15}$$

The case $x_B = x$ does not occur when the slab is perpendicular to the x-axis. The y- and z- coordinates of the point P_n have to be found in order to determine whether the ray "hits" the slab, which may be of limited dimensions.

$$x_n \equiv R_n \tag{6-16}$$

$$y_n = y + (x_n - x)\frac{y_B - y}{x_B - x} \tag{6-17}$$

$$z_n = z + (x_n - x)\frac{z_B - z}{x_B - x} \tag{6-18}$$

6.1.3. Slab Shield Perpendicular to the Cylinder Axis

Figure 6.4 shows a projection on the (y,z) plane of this case. The shield has the thickness L_n and for the plane facing the source $z = z_n$. Often $z_n = 0$, i.e. the shield is in contact with the source. The distance travelled in the shield is

$$d_n = \frac{R_B}{|z_B - z|} L_n, \tag{6-19}$$

and the distance travelled inside the source is

$$d_s = \frac{R_B}{|z_B - z|}\, z. \tag{6-20}$$

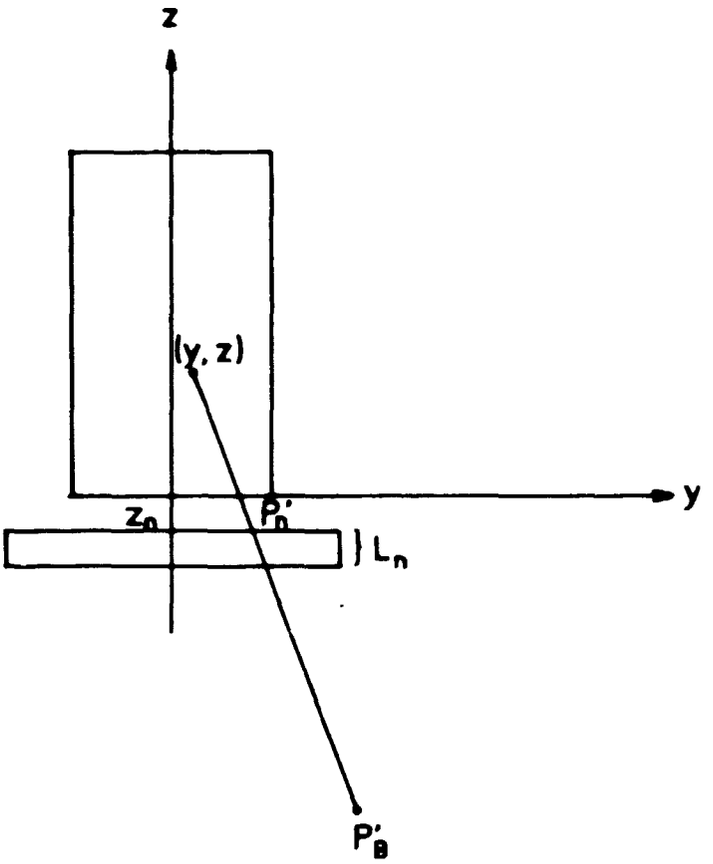

Fig. 6.4. Projection on (y,z)-plane. Slab shield perpendicular to the cylinder axis.

Figures 6.5 and 6.6 illustrate a situation where the case treated above is used. The tube with a 90° bend can be treated by superposition of the two cylindrical sources A and B, where A has a cylindrical shield plus the additional slab shield parallel to its axis and B has two slab shields perpendicular to its axis - plus a cylindrical shield, which, however, is of no interest with the observation point placed as shown in the figures.

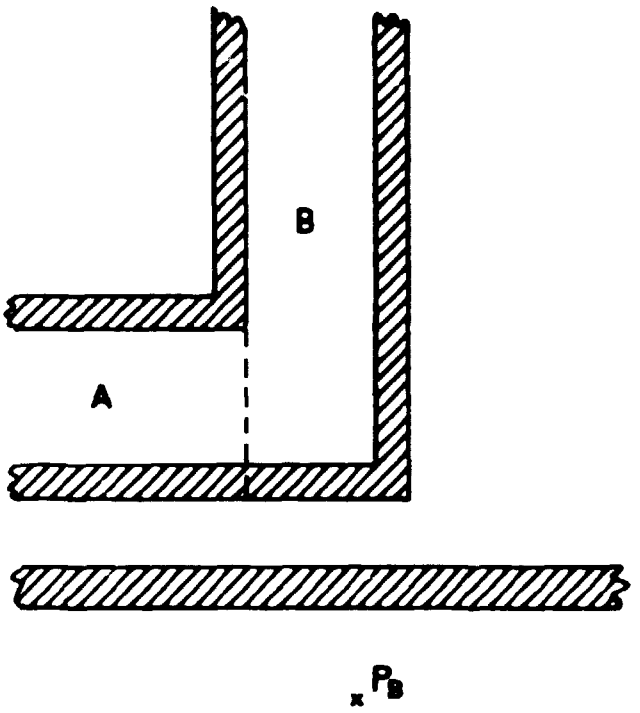

Fig. 6.5. Tube with a 90°-bend and an additional shielding layer.

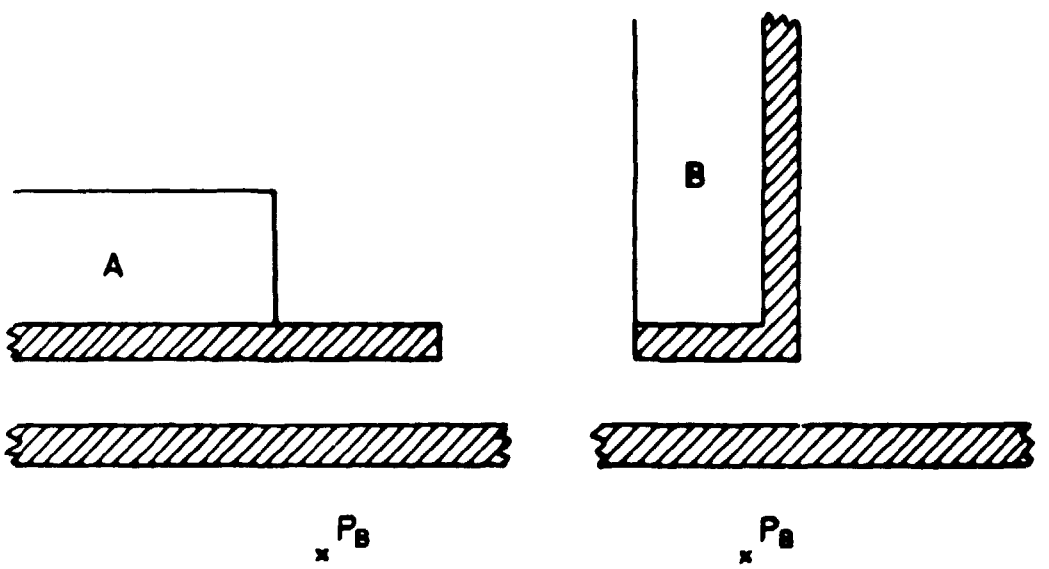

Fig. 6.6. Dissolution of the case in fig. 6.5.

6.1.4. Evaluation of the Integrals

The integrals in eqs. (6-2) and (6-3) are evaluated numerically by Gaussian integration. With this method an integral going from -1 to +1 is converted into a sum of weighted functional values:

$$\int_{-1}^{1} f(x)dx = \sum_{i=1}^{n} w_i f(x_i) \qquad (6\text{-}21)$$

where

n = the order of the method, $n \geq 2$,
w_i = the weight factors, and
x_i = the abscissas, where the functional values are taken.

The order is often chosen as n = 7 or n = 20, which in most cases give sufficiently good results. The abscissas x_i are zeros of Legendre polynomials. These and the weight factors are available in many standard mathematical tables and also as standard computer programs.

The integrals in (6-2) and (6-3) are now converted into forms with limits -1 and +1.

6.1.4.1. Volume Source. Rewriting (6-2) and omitting (r,θ,z) gives:

$$\Phi_B = 3.7 \cdot 10^{10} \int_{z=0}^{H} \int_{\theta=0}^{2\pi} \int_{r=0}^{R} S_V B \, e^{-\sum \mu_n d_n} \frac{1}{4\pi R_B^2} \, r \, dr d\theta dz \qquad (6\text{-}22)$$

The following substitutions are made:

$$r' \equiv \frac{2}{R}r - 1 \Rightarrow r = \tfrac{1}{2}R(r'+1) \Rightarrow dr = \tfrac{1}{2}Rdr' \quad , \qquad (6\text{-}23)$$

$$\theta' \equiv \frac{1}{\pi}\theta - 1 \Rightarrow \theta = \pi(\theta'+1) \Rightarrow d\theta = \pi d\theta' \quad , \qquad (6\text{-}24)$$

$$z' \equiv \frac{2}{H}z - 1 \Rightarrow z = \tfrac{1}{2}H(z'+1) \Rightarrow dz = \tfrac{1}{2}Hdz' \quad . \qquad (6\text{-}25)$$

Hereby the marked variables will go from -1 to +1 and (6-22) becomes

$$\Phi_B = 3.7\cdot10^{10}\,\frac{\pi}{4}\,RH\int_{-1}^{1}\int_{-1}^{1}\int_{-1}^{1} S_V B\; e^{-\sum_n \mu_n d_n}\frac{1}{4\pi R_B^2} r\,dr'\,d\theta'\,dz' \qquad (6\text{-}26)$$

which by means of (6-21) is converted into

$$\Phi_B = 3.7\cdot10^{10}\,\frac{\pi}{4}\,RH\sum_{i=1}^{n_i}\sum_{j=1}^{n_j}\sum_{k=1}^{n_k} f(r_k',\theta_j',z_i')\,w_k w_j w_i \qquad (6\text{-}27)$$

where

$$f(r_k',\theta_j',z_i') \equiv S_V(r_k,\theta_j,z_i)B(r_k,\theta_j,z_i)\exp(-\Sigma\mu_n d_n(r_k,\theta_j,z_i)) \cdot \frac{r_k}{4\pi R_B(r_k,\theta_j,z_i)^2} \; . \qquad (6\text{-}28)$$

As indicated in (6-27), the three Gaussian integrations need not be of the same order, but of course they often are.

6.1.4.2. Surface Source. Equation (6-3) becomes

$$\Phi_B = 3.7\cdot10^{10}\int_{z=0}^{H}\int_{\theta=0}^{2\pi} S_S B\; e^{-\sum_n \mu_n d_n}\frac{1}{4\pi R_B^2} R\,d\theta\,dz \qquad (6\text{-}29)$$

$$= 3.7\cdot10^{10}\,\frac{\pi}{2}\,RH\int_{-1}^{1}\int_{-1}^{1} S_S B\; e^{-\sum_n \mu_n d_n}\frac{1}{4\pi R_B^2}\,d\theta'\,dz' \qquad (6\text{-}30)$$

using the substitutions (6-24) and (6-25). The conversion into sums takes place as above:

$$\Phi_B = 3.7\cdot10^{10}\,\frac{\pi}{2}\,RH\sum_{i=1}^{n_i}\sum_{j=1}^{n_j} f(\theta_j',z_i')\,w_j w_i \qquad (6\text{-}31)$$

where

$$f(\theta_j',z_i') \equiv S_S(\theta_j,z_i)B(\theta_j,z_i)\exp(-\Sigma_n \mu_n d_n(\theta_j,z_i)\,\frac{1}{4\pi R_B(\theta_j,z_i)^2} \qquad (6\text{-}32)$$

6.2. Evaluation of Build-Up Factors and Absorption Coefficients

The dose build-up factor B is an "effective build-up factor" for a multilayer shield. It is difficult to determine a proper combination of the single build-up factors as these are dependent upon energy as well as the number of relaxation lengths. Thus the order of the different layers becomes important for which formula to use. The first layer is the source itself, i.e. steam or water, and the second layer is the steel tubing or casing. As it can be assumed that there is no build-up in steam, this does not give any problems. For water, however, the build-up factor must be combined with that for steel. In accordance with [22], the total build-up factor is evaluated as

$$B(E,\mu_1 d_1,\mu_2 d_2) = B_2(E,\mu_1 d_1 + \mu_2 d_2) \tag{6-33}$$

where B_2 is the build-up factor of the second layer (iron). If there are additicnal layers of shielding, the total build-up factor is found as the product of the build-up factors of these layers and expression (6-33):

$$B(E,\mu_1 d_1,\mu_2 d_2,\ldots,\mu_n d_n) = B_2(E,\mu_1 d_1+\mu_2 d_2) \prod_{p=3}^{n} B_p(E,\mu_p d_p) \ . \tag{6-34}$$

It must be stressed that approximation (6-34) becomes poorer the more layers there are, so it must be used with care. In most cases, however, there is not more than one extra layer of shielding so that the total build-up factor should be reasonably close to reality.

The single layer build-up factors are calculated by a polynomial approximation as functions of energy, E, and relaxation lengths $\mu_n d_n$ [23]

$$B(E,\mu_n d_n) = \sum_{j=0}^{4} \sum_{i=0}^{3} C_{ij} \cdot (\mu_n d_n)^i \cdot \left(\frac{1}{E}\right)^j \ . \tag{6-35}$$

Values of the polynomial coefficients C_{ij} for water, iron, and three types of concrete have been taken from [23,24].

The absorption coefficients used in the calculations are derived by polynomial approximations to values taken from tables in [25]. Hereby the mass absorption coefficient, μ/ρ, which is the one tabulated, can be expressed by

$$\frac{\mu}{\rho} = 10^{P(E)} \tag{6-36}$$

where P is a polynomial in the energy E:

$$P(E) = A_0 + A_1 \log_{10}E + A_2 (\log_{10}E)^2 + ... + A_{10} (\log_{10}E)^{10}$$

$$= \sum_{i=0}^{10} A_i (\log_{10}E)^i \ . \tag{6-37}$$

The polynomial coefficients A_i are found by fitting (6-36) to tabulated values of $\mu(E)/\rho$.

6.3. Conversion of Units

Equation (6-1) gives the dose rate at point P_B in terms of MeV/g·s. Often the unit rad/h will be preferred.

$$\dot{D}_B \ \text{rad/h} = 5.76 \cdot 10^{-5} \ \dot{D}_B \ \text{MeV/g·s} \ . \tag{6-38}$$

The exposure rate at the same point is

$$\dot{X}_B \ \text{R/h} = \frac{1}{0.87} \ \dot{D}_B \ \text{rad/h} \ . \tag{6-39}$$

A person standing at P_B will be subjected to a dose rate

$$\dot{D} \simeq 0.95 \ \dot{X} \ \text{(rad/h)} \ . \tag{6-40}$$

The quality factor for gamma radiation equals 1, so the rate of dose equivalent received by the person is

$$\dot{H} = 1 \cdot \dot{D} \ \text{(rem/h)} \tag{6-41}$$

6.4. The Importance of Knowing the Exact Measurements of the Source

A problem that often arises is lack of knowledge of the length and radius of the source. Especially the length is rarely given in safety analysis reports. In order to judge the order of magnitude of the error made by incorrectly guessing the value of one of these measurements, a small parameter study was performed. Figure 6.7 shows the arrangement. The source is shielded by a

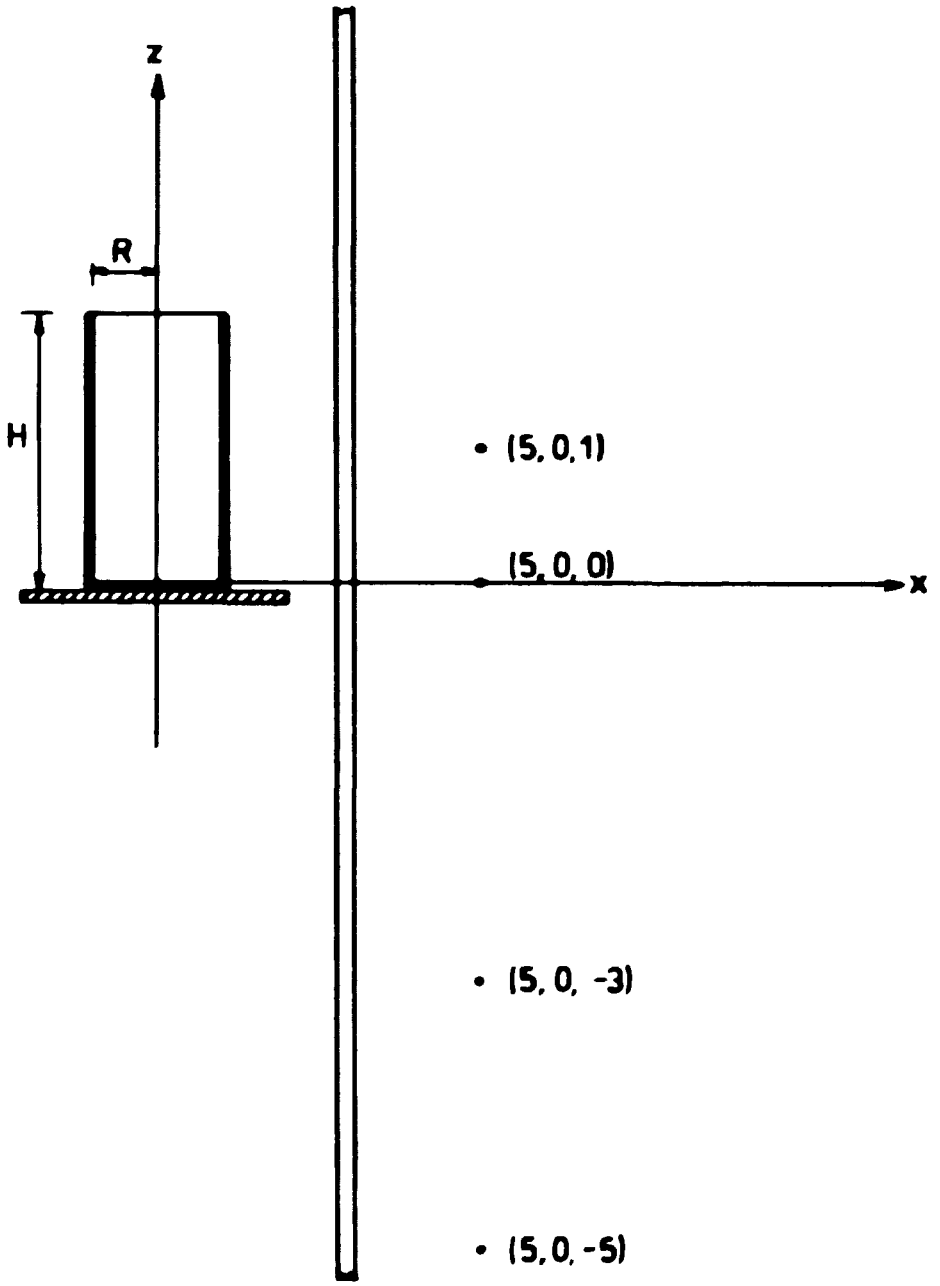

Fig. 6.7. Test case for parameter study.

5 cm thick steel tube and a 10 cm concrete slab. At the one end the shield consists of 5 cm of steel and 5 cm of concrete. The source contains 1 Ci of a nuclide emitting 1 MeV photons. The exposure rate is calculated in four points as indicated in the figure. Figure 6.8 shows the effect of varying the radius between 5 and 25 cm, while keeping the length at 2 m, and of varying the length between 1 and 10 m with constant radius equal 25 cm. As seen, even a very bad guess for the radius will not result in a change of the exposure rate of more than a factor of about 2. This is also the case for the two observation points above z=0 when the length is changed, while for points below z=0 greater variations may occur.

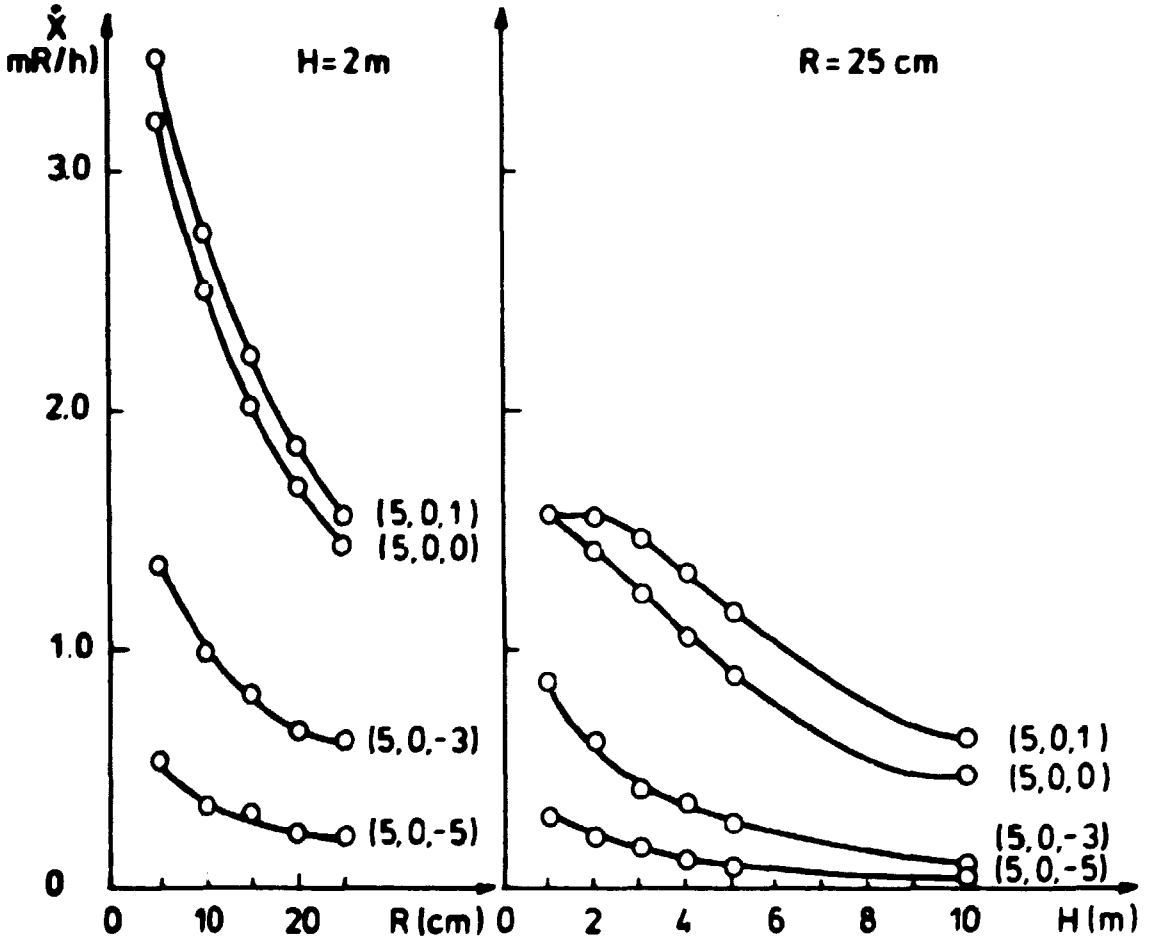

Fig. 6.8. Result of parameter study.

7. COMPUTER PROGRAMMING

The mathematical models described in the preceding chapters have been programmed in FORTRAN IV. Figure 7.1 shows a block diagram of the procedure for calculating exposure rates and

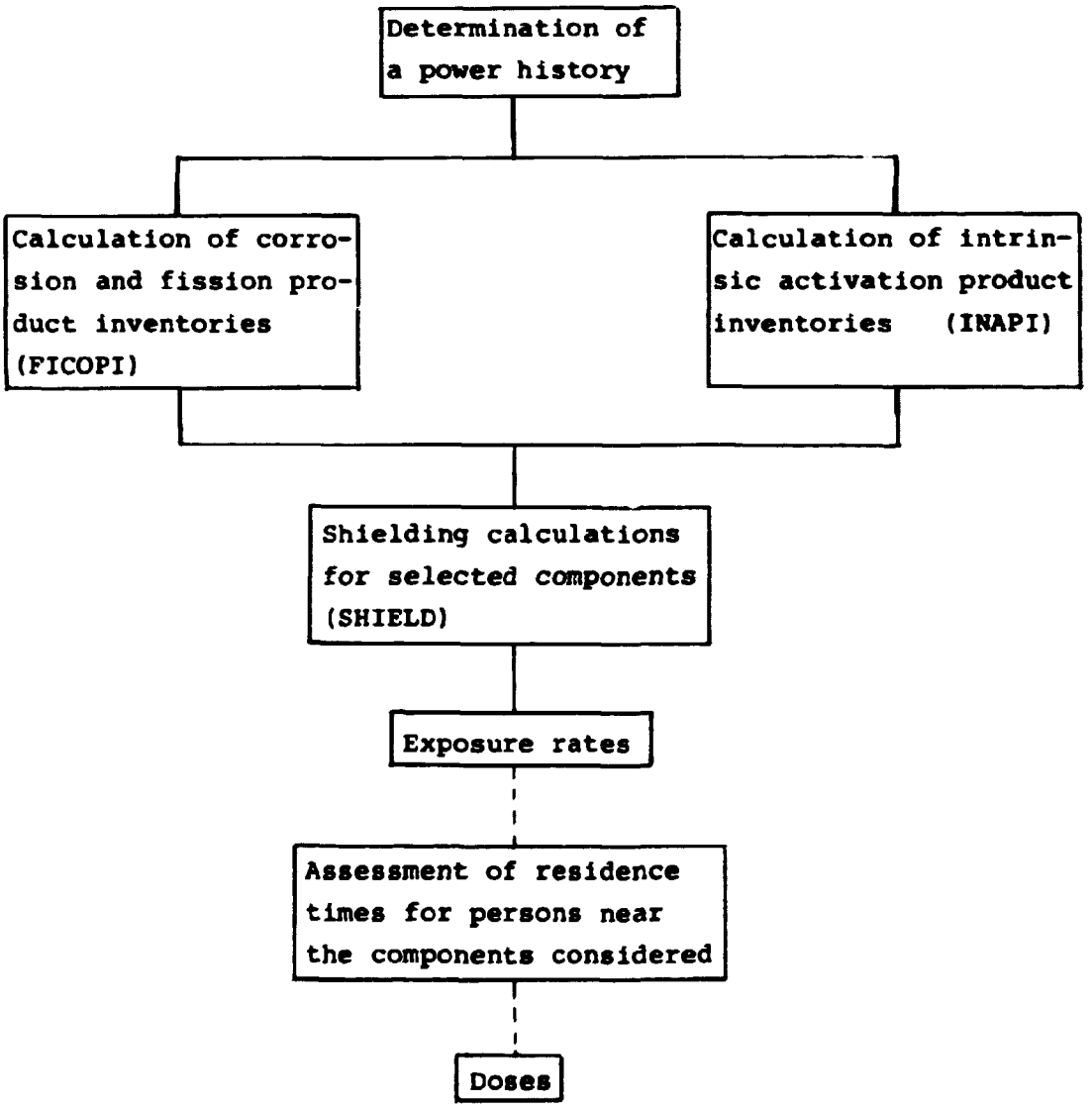

Fig. 7.1. Block diagram for the calculation of exposure rates and doses.

doses. The dotted lines indicate that this point has not been studied in greater detail. So the calculations cease at exposure rates. Two major codes were developed: FICOPI (Fission and Corrosion Product Inventories) and SHIELD. The code INAPI (Intrinsic Activation Product Inventories) is a small one that only integrates eq. (5-3). A closer description of the codes will not be given here. Only the central parts of FICOPI are outlined in figs. 7.2 and 7.3. Figure 7.2 shows a block diagram of the main program, which essentially has the task of introducing the power history into the program, i.e. governing the time for and duration of operational and shutdown periods, respectively. Figure 7.3 shows a simplified block diagram for SUBROUTINE EULER, which is the subroutine solving the differential equations during operation, i.e. eqs. (3-1) and (3-11). As shown in section 3.1.2, the system of differential equations is turned into a system of algebraic equations for each time-step, which again is split into NJ systems of equations where NJ is the number of nuclides in the decay chain. As seen from eq. (3-19), the coefficient matrices of the system only change when the time increment is changed. This fact is utilized in the subroutine and causes a substantial decrease in computer time.

The cpu-time for a run with FICOPI is, roughly speaking, proportional to the number of operational periods and to NJ, the number of nuclides in the decay chain under consideration. The shutdown periods consume little cpu-time compared to the operational periods. On the Burroughs B-6700 computer a run over one operational period with one single nuclide takes about 30 s cpu-time. The time constants of the specific problem have some influence on this number and corrosion product calculations tend to be more time-consuming than fission product calculations, taking typically 80-90 s cpu-time for a run with two nuclides over one operational period.

The cpu-time consumed by the shielding code SHIELD is small compared to the time for FICOPI. The order of magnitude is 1 s to compute the exposure rate at one point from a cylinder source with two shielding layers.

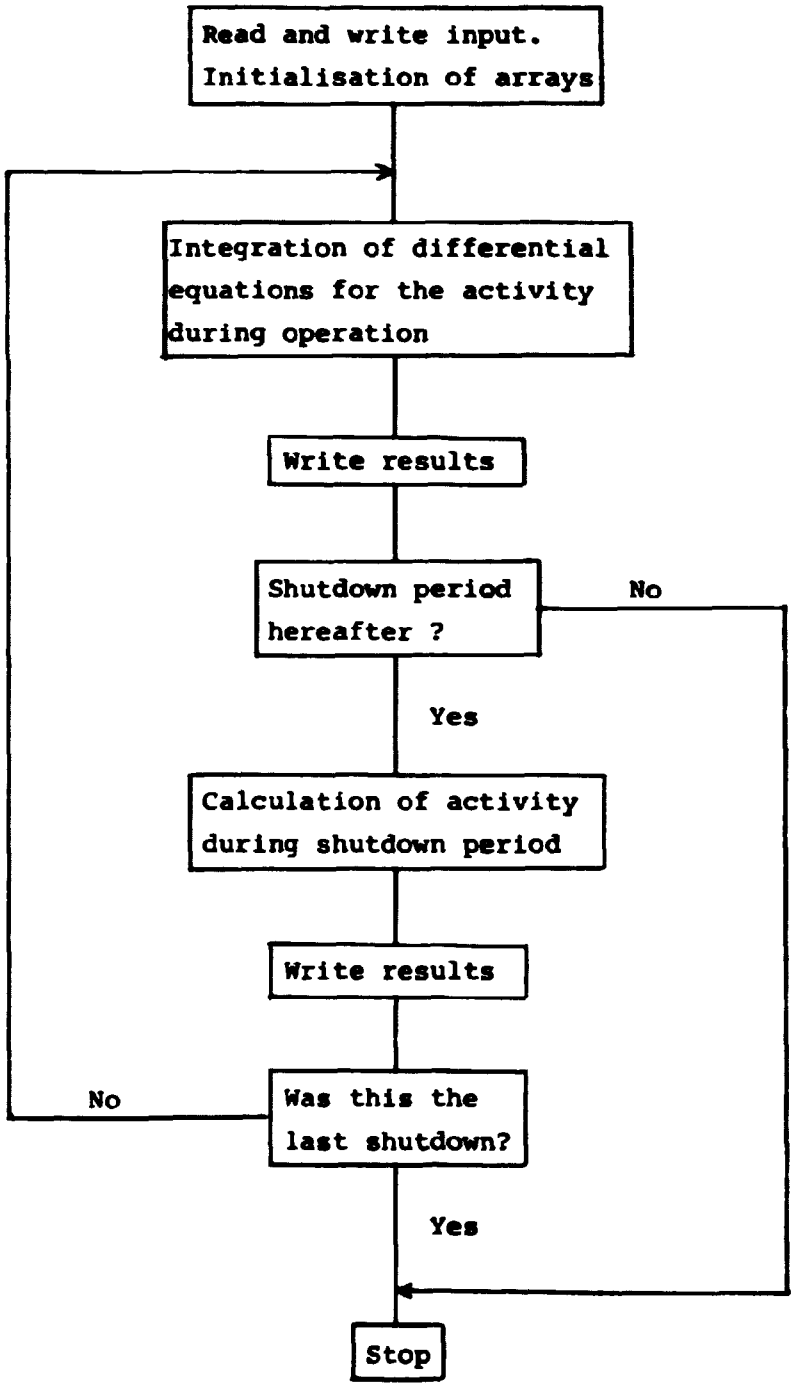

Fig. 7.2. Block diagram of FICOFI, main program.

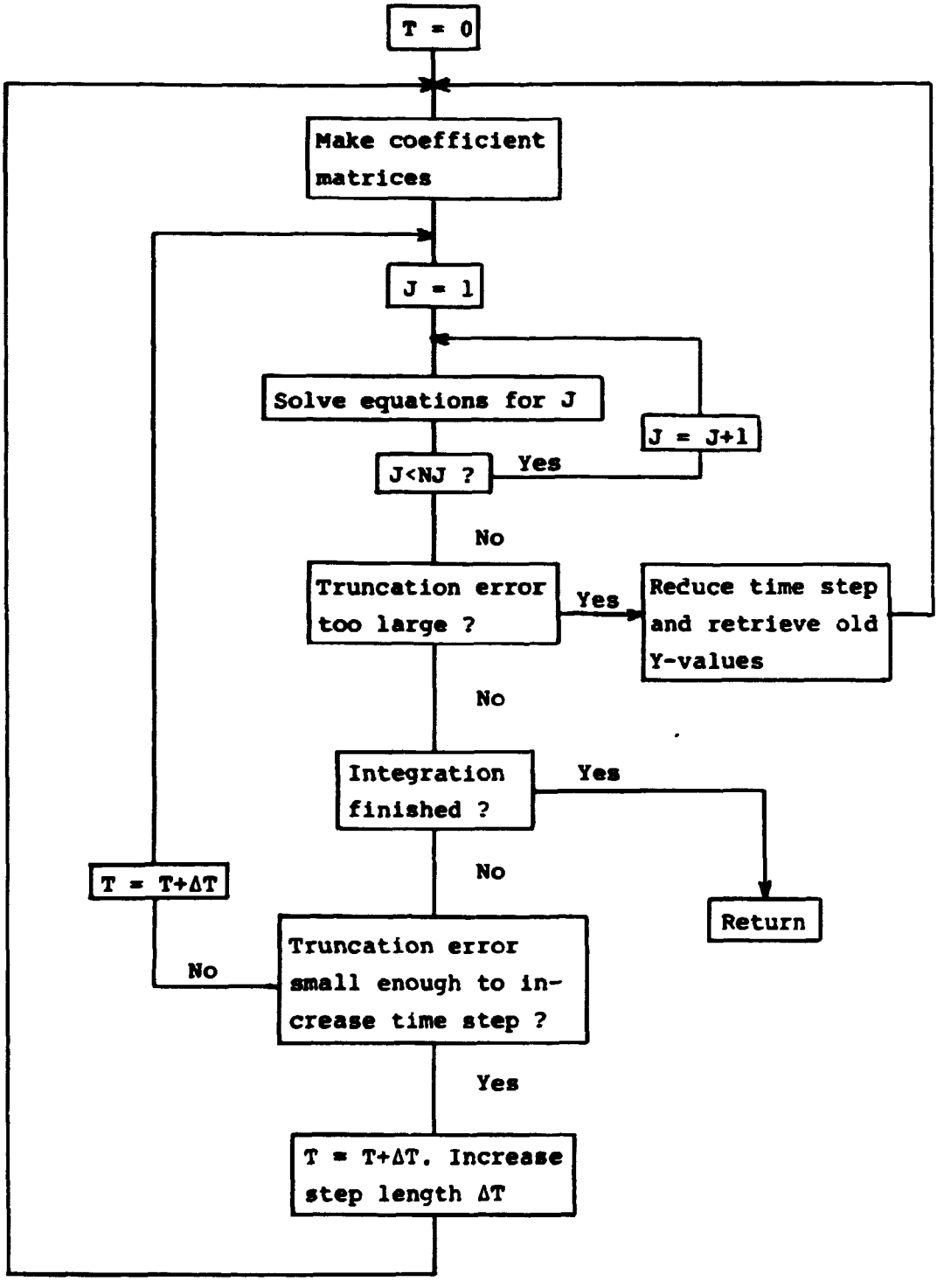

Fig. 7.3. Block diagram of FICOP1, SUBROUTINE EULER.

8. CALCULATIONS PERFORMED WITH THE CODES

In order to test the codes against measurements and other calculations, a test case was run. Figure 8.1 shows the idealised power history applied. It starts with a period of 1½ years at full power followed by 2 months shutdown. After this comes

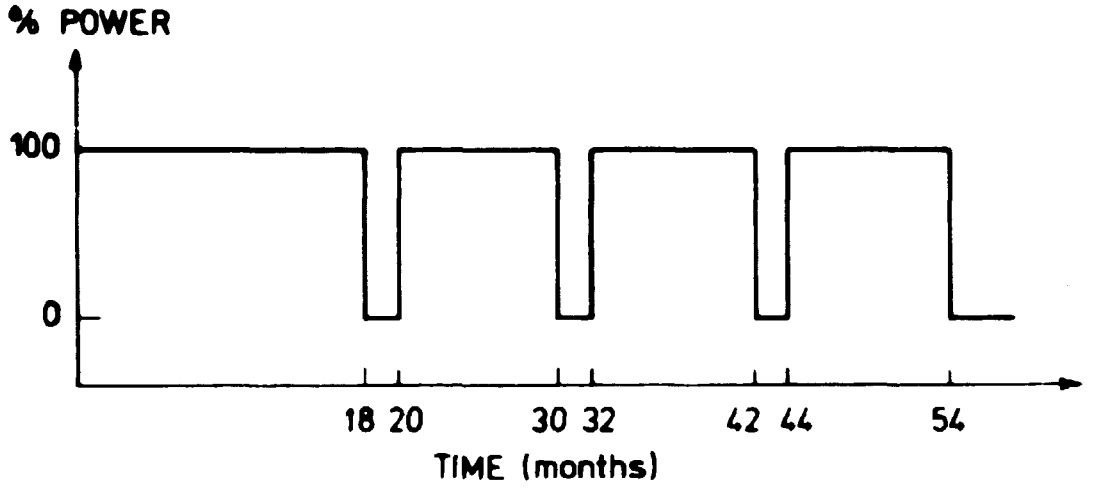

Fig. 8.1. Power history.

three periods of 10 months at full power separated by shutdown periods lasting 2 months each. In table 8.1 are listed a number of the plant data used for input in the codes. Most of these data have been taken from [1], while due to lack of information others are more or less guessed values. Values for inventory change rates and partition factors are not given in the table due to their large number; they are found in [1].

The results of the calculations have been compared to data given in [1], which are partly results of calculations, partly measured data from different General Electric BWRs.

The fission product inventories during operation after 1½ years calculated by FICOPI for most of the nuclides studied are in good agreement (within a factor 5) with values given in [1]. However, a number of nuclides show great disagreement. The reason for these deviations has not been revealed yet, but possible sources of errors are: partition factors and release rates

Table 8.1

Some plant data used in the calculations

Thermal power	3579 MW	
Electrical power	1250 MW	
Neutron fluxes		
Thermal ($\leq$ 0.625eV)	$2.9 \cdot 10^{13}$	$cm^{-2}s^{-1}$
> Thermal	$2.05 \cdot 10^{14}$	$cm^{-2}s^{-1}$
Coolant inventories		
Reactor core	$3.936 \cdot 10^{4}$	kg
Downcomer	$9.096 \cdot 10^{4}$	-
Recirculation system (2 loops)	$1.985 \cdot 10^{4}$	-
Lower plenum	$7.908 \cdot 10^{4}$	-
Reactor water clean-up system	$0.100 \cdot 10^{4}$	-
Upper plenum and separators	$5.295 \cdot 10^{4}$	-
Total in reactor water components	$2.832 \cdot 10^{5}$	kg
Main steam lines (4 lines)	$4.052 \cdot 10^{3}$	kg
High pressure turbine	170	kg
Mass flow rates at full power		
Through core	13230	kg/s
To turbine	1940	-
In recirculation system	3364	-
Flow area in core	7.32	m^2
Outside diameter of steam line	0.66	m
Wall thickness of steam line	0.05	m
Steam volume in steam lines	113.2	m^3
Length of high pressure turbine (HPT)	4.00	m
Wall thickness of HPT	0.05	m
"Equivalent diameter" of HPT	1.22	m
Deposition and release rate constants:		
For noble gases and halogens		
Deposition	0.0	s^{-1}
Release	0.0	s^{-1}
For other elements		
Deposition	$1.3 \cdot 10^{-5}$	s^{-1}
Release	$5 \cdot 10^{-8}$	s^{-1}

from fuel. Table 8.2 lists the inventories of some of the more important nuclides in the High Pressure Turbine as calculated by FICOPI and as given in [1].

Table 8.2

Comparison of activity inventories for HPT

Nuclide	FICOPI	Ref. 1
^{89}Kr	$1.1 \cdot 10^{-2}$ Ci	$3.7 \cdot 10^{-3}$ Ci
^{90}Kr	$2.1 \cdot 10^{-2}$ Ci	$6.1 \cdot 10^{-4}$ Ci
^{135m}Xe	$4.9 \cdot 10^{-3}$ Ci	$2.4 \cdot 10^{-3}$ Ci
^{135}Xe	$2.0 \cdot 10^{-3}$ Ci	$1.1 \cdot 10^{-3}$ Ci
^{137}Xe	$1.4 \cdot 10^{-2}$ Ci	$5.5 \cdot 10^{-3}$ Ci
^{139}Xe	$2.2 \cdot 10^{-2}$ Ci	$8.7 \cdot 10^{-4}$ Ci

In the case of activated corrosion products, it was only possible to compare with data for the concentration in reactor water. These data are also from [1] and are average values of measurements from different plants. Table 8.3 compares results from FICOPI with these data, showing quite good agreement taking into consideration the uncertainties concerning corrosion and release rates.

Table 8.3

Corrosion product concentrations in reactor water

Nuclide	FICOPI	Ref. 1
^{51}Cr	$2.0 \cdot 10^{-6}$ Ci/kg	$5 \cdot 10^{-7}$ Ci/kg
^{54}Mn	$1.6 \cdot 10^{-7}$ Ci/kg	$4 \cdot 10^{-8}$ Ci/kg
^{56}Mn	$1.3 \cdot 10^{-6}$ Ci/kg	$5 \cdot 10^{-5}$ Ci/kg
^{58}Co	$1.8 \cdot 10^{-6}$ Ci/kg	$5 \cdot 10^{-6}$ Ci/kg
^{60}Co	$2.6 \cdot 10^{-9}$ Ci/kg	$5 \cdot 10^{-7}$ Ci/kg
^{59}Fe	$2.5 \cdot 10^{-8}$ Ci/kg	$8 \cdot 10^{-8}$ Ci/kg

Calculations of the radiation levels near components were performed for the main steam lines and for the high pressure

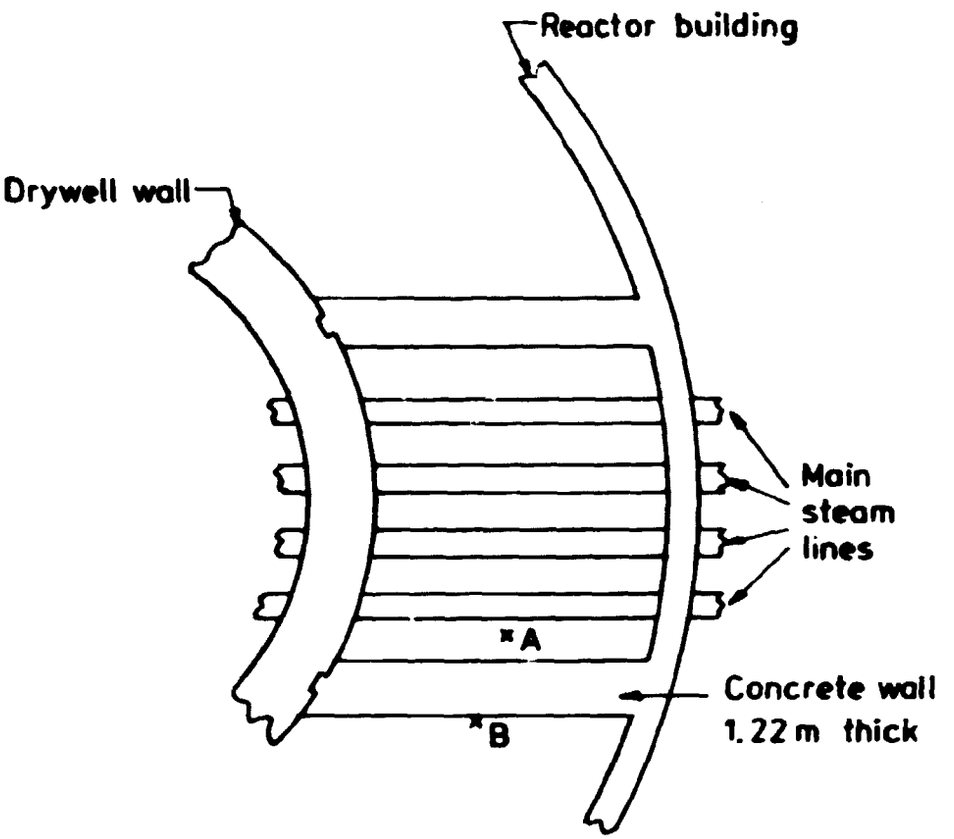

Fig. 8.2. Main steam lines.

turbine. Figure 8.2 shows a sketch of the arrangement where the main steam lines penetrate the drywell and the reactor building. The lengths of the pipe sections considered are about 6 m. The radiation field was calculated for points A and B, respectively. Point A lies at a distance of one radius (= 28 cm) from the nearest pipe, and point B lies on the outer surface of the concrete wall. Table 8.4 shows the results of the calculations. As expected, ^{16}N dominates the radiation field during operation. In [1] measured levels on main steam lines of 1.5-2.0 R/h are reported, so there seems a reasonable agreement. The shutdown level, which is very low, is due to the nuclides ^{137}Cs, ^{54}Mn, and ^{58}Co.

Table 8.4

Radiation levels (R/h) from main steam lines

Location	Operation		After 2 months shutdown	
	^{16}N	Others	^{16}N	Others
A	1.0	$4.5 \cdot 10^{-3}$	0	$\sim 4.1 \cdot 10^{-7}$
B	$1.22 \cdot 10^{-4}$	$\sim 10^{-8}$	0	0

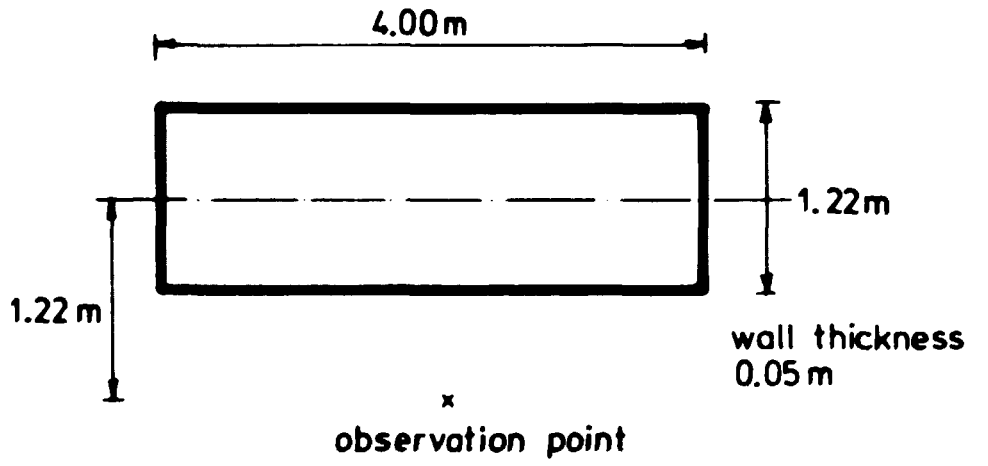

Fig. 8.3. High pressure turbine.

Figure 8.3 shows the arrangement of the high pressure turbine. As the measurements of the turbine are not given in [1], a length of 4 m was guessed. This is a normal length for the high pressure part of a 1500 r/min turbine for nuclear power plants. The "equivalent diameter" is the diameter of a cylinder of length 4 m and with a volume as given in [1], cf. table 8.1. This diameter is used for the shielding calculation. Here, too, the dominant nuclide during operation is ^{16}N. The level of 0.594 R/h should be compared to measured levels of 0.150-0.7 R/h as reported in [1]. The shutdown radiation is due to ^{137}Cs, ^{54}Mn, and ^{58}Co.

Table 8.5

Radiation levels (R/h) from the high pressure turbine

Operation		After 2 months' shutdown	
^{16}N	Others	^{16}N	Others
0.594	$7.0 \cdot 10^{-3}$	0	$4.0 \cdot 10^{-7}$

9. CONCLUSION

A model was set up with the purpose of calculating the radioactive inventories of components in the coolant circuit of a nuclear power plant with a boiling water reactor. Furthermore, a simple model for the calculation of the shielding was implemented in order to calculate radiation fields outside the components.

Emphasis was laid on the modelling of the transport of radioactive material outside the reactor. The radiation field during operation around the reactor itself due to neutrons, fission gammas, etc., was not investigated. One reason for this is that the areas around the reactor, where these sources might be of importance, are not accessible during operation, so that this radiation does not contribute to personnel doses.

The power plant model describes the various components as "black boxes", which are ascribed a number of parameters such as inventory change rate and partition factors. Hereby the total amount of a nuclide in the coolant and on the walls of the components can be evaluated, but the distribution of the activity within one component cannot be found. This may be a disadvantage for components with complex geometries such as turbines, but probably does not matter for tubes and similar simple geometries.

Some calculations of inventories and radiation fields were performed in order to test the model. The results show reasonable agreement with available data from measurements or calculations using other models. However, the number of comparisons made so far allow no evaluation of the validity of the model as a whole. A major drawback of a model of this kind is its dependence on rather unknown data such as, for instance, the erosion

and deposition rates, or on data that are rarely available in the open literature, e.g. geometrical measurements of power plant components.

Provided all the necessary data are reasonably well known, the model should be a useful tool in the assessment of radiation fields. If, furthermore, it is possible to predict the residence time for persons in locations where there is radiation, the annual man-rem dose for a power plant could be calculated. However, it is more likely that the usefulness of such a model lies in the possibility for studying the influence on radiation fields of various parameters such as, for instance, the choice of construction materials.

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APPENDIX A

Derivation of Expressions for Inventories During Shutdowns

In this appendix a thorough evaluation of the analytical expressions used in section 3.2.2 for inventories is given. The index i for component number is omitted for convenience, where this cannot lead to misinterpretations.

Turbine System Components. Equations (3-25) and (3-26) for $n_{ij}(t)$ and $w_{ij}(t)$ are of the same form, so only (3-25) is treated here. This equation represents a series of equations for the nuclides in a decay chain:

$$\frac{d\, n_1(t)}{dt} = - \lambda_1 n_1(t) \tag{A-1}$$

$$\frac{d\, n_2(t)}{dt} = \lambda_1 b_{1,2} n_1(t) - \lambda_2 n_2(t) \tag{A-2}$$

$$\vdots$$

$$\frac{d\, n_j(t)}{dt} = \lambda_{j-1} b_{j-1,j} n_{j-1}(t) - \lambda_j n_j(t) \tag{A-3}$$

The solutions for n_1 and n_2 are:

$$n_1(t) = n_1(0) e^{-\lambda_1 t} \tag{A-4}$$

$$n_2(t) = e^{-\lambda_2 t} \left\{ \int \lambda_1 b_{1,2} n_1(0) e^{-\lambda_1 t} e^{\lambda_2 t}\, dt + C_2^n \right\}$$

$$= \frac{\lambda_1 b_{1,2} n_1(0)}{\lambda_2 - \lambda_1} e^{-\lambda_1 t} + C_2^n e^{-\lambda_2 t} \tag{A-5}$$

where

$$C_2^n = n_2(0) - \frac{\lambda_1 b_{1,2}}{\lambda_2 - \lambda_1} n_1(0) \tag{A-6}$$

It is now postulated that the solution for $n_j(t)$ is

$$n_j(t) = \sum_{k=1}^{j-1} \left(\prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \right) C_k^n e^{-\lambda_k t} + C_j^n e^{-\lambda_j t} \qquad \text{(A-7)}$$

$$C_j^n = n_j(0) - \sum_{k=1}^{j-1} \left(\prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \right) C_k^n \qquad \text{(A-8)}$$

To prove that (A-7) is valid for all j-values greater than or equal to 2, one must prove that it is valid for j = 2, and that it is valid for j + 1 as a consequence of being valid for j. A comparison between expressions (A-5) and (A-7) easily shows that the first condition is fulfilled. With respect to the second one:

$$n_{j+1}(t) = e^{-\lambda_{j+1} t} \left\{ \int \lambda_j b_{j,j+1} n_j(t) e^{\lambda_{j+1} t} dt + C_{j+1}^n \right\}$$

$$= e^{-\lambda_{j+1} t} \left\{ \int \lambda_j b_{j,j+1} \left(\sum_{k=1}^{j-1} \left(\prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \right) C_k^n e^{-\lambda_k t} + C_j^n e^{-\lambda_j t} \right) e^{\lambda_{j+1} t} dt + C_{j+1}^n \right\}$$

$$= e^{-\lambda_{j+1} t} \lambda_j b_{j,j+1} \sum_{k=1}^{j-1} \left(\prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \right) C_k^n \int e^{(\lambda_{j+1} - \lambda_k) t} dt$$

$$+ \lambda_j b_{j,j+1} e^{-\lambda_{j+1} t} C_j^n \int e^{(\lambda_{j+1} - \lambda_j) t} dt + C_{j+1}^n e^{-\lambda_{j+1} t}$$

$$= \sum_{k=1}^{j-1} \lambda_j b_{j,j+1} \left(\prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \right) C_k^n \frac{e^{-\lambda_k t}}{\lambda_{j+1} - \lambda_k}$$

$$+ \frac{\lambda_j b_{j,j+1}}{\lambda_{j+1} - \lambda_j} C_j^n e^{-\lambda_j t} + C_{j+1}^n e^{-\lambda_{j+1} t}$$

$$= \sum_{k=1}^{j-1} \left(\prod_{p=k}^{j} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \right) C_k^n e^{-\lambda_k t} + \frac{\lambda_j b_{j,j+1}}{\lambda_{j+1} - \lambda_j} C_j^n e^{-\lambda_j t}$$

$$+ C_{j+1}^{n} e^{-\lambda_{j+1} t}$$

$$= \sum_{k=1}^{j} \left(\prod_{p=k}^{j} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \right) C_k^n e^{-\lambda_k t} + C_{j+1}^{n} e^{-\lambda_{j+1} t}$$

q.e.d.

$$C_{j+1}^{n} = n_{j+1}(0) - \sum_{k=1}^{j} \left(\prod_{p=k}^{j} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \right) C_k^n$$

which is the same form as (A-8).

By defining

$$K_{kj} \equiv \prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\lambda_{p+1} - \lambda_k} \qquad \text{for } k \leq j-1$$

$$K_{jj} \equiv 1$$

we obtain the expression used in (3-29):

$$n_{ij}(t) = \sum_{k=1}^{j} K_{kj} C_k^n e^{-\lambda_k t}$$

Reactor Water. The differential equation (3-36) is very similar to (3-25) for the turbine system, the main difference being the constant source term, S_j. Thus the solution is derived in the same way as above. Equation (3-36) represents a series of differential equations:

$$\frac{dn_1(t)}{dt} = S_1 - \beta_1 n_1(t) \qquad \text{(A-9)}$$

$$\frac{dn_2(t)}{dt} = S_2 + \lambda_1 b_{1,2} n_1(t) - \beta_2 n_2(t) \qquad \text{(A-10)}$$

$$\vdots$$

$$\frac{dn_j(t)}{dt} = S_j + \lambda_{j-1} b_{j-1,j} n_{j-1}(t) - \beta_j n_j(t) \qquad \text{(A-11)}$$

Solutions for n_1 and n_2 are

$$n_1(t) = e^{-\int \beta_1 dt}\left\{\int S_1 e^{\int \beta_1 dt} dt + C_1\right\}$$

$$= \frac{1}{\beta_1} S_1 + C_1 e^{-\beta_1 t} \qquad \text{(A-12)}$$

$$C_1 = n_1(0) - \frac{1}{\beta_1} S_1$$

$$n_2(t) = e^{-\int \beta_2 dt}\left\{\int (S_2 + \lambda_1 b_{1,2}(\frac{1}{\beta_1} S_1 + C_1 e^{-\beta_1 t}))e^{\int \beta_2 dt} dt + C_2\right\}$$

$$= \frac{1}{\beta_2} S_2 + \frac{1}{\beta_2} \frac{\lambda_1}{\beta_1} b_{1,2} S_1 + \frac{\lambda_1 b_{1,2}}{\beta_2 - \beta_1} C_1 e^{-\beta_1 t} + C_2 e^{-\beta_2 t} \qquad \text{(A-13)}$$

$$C_2 = n_2(0) - \frac{1}{\beta_2}(S_2 + \frac{\lambda_1}{\beta_1} b_{1,2} S_1) - \frac{\lambda_1 b_{1,2}}{\beta_2 - \beta_1} C_1 \qquad \text{(A-14)}$$

It is now postulated that $n_j(t)$ for $j \geq 2$ follows the expression

$$n_j(t) = \frac{1}{\beta_j} S_j + \frac{1}{\beta_j} \sum_{k=1}^{j-1} S_k \left(\prod_{p=k}^{j-1} \frac{\lambda_p}{\beta_p} b_{p,p+1}\right)$$

$$+ \sum_{k=1}^{j-1} \left(\prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\beta_{p+1} - \beta_k}\right) C_k e^{-\beta_k t} + C_j e^{-\beta_j t} \qquad \text{(A-15)}$$

$$C_j = n_j(0) - \frac{1}{\beta_j} S_j - \frac{1}{\beta_j} \sum_{k=1}^{j-1} S_k \left(\prod_{p=k}^{j-1} \frac{\lambda_p}{\beta_p} b_{p,p+1}\right)$$

$$- \sum_{k=1}^{j-1} \left(\prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\beta_{p+1} - \beta_k}\right) C_k \qquad \text{(A-16)}$$

As is easily verified, $n_2(t)$ follows this expression, and it is now to be proved that $n_{j+1}(t)$ follows it when $n_j(t)$ does.

$$n_{j+1}(t) = e^{-\int \beta_{j+1} dt} \left\{ \int (S_{j+1} + \lambda_j b_{j,j+1} n_j(t)) e^{\int \beta_{j+1} dt} dt + C_{j+1} \right\}$$

$$= \frac{1}{\beta_{j+1}} S_{j+1} + e^{-\beta_{j+1} t} \left\{ \lambda_j b_{j,j+1} \int \left(\frac{1}{\beta_j} S_j \right.\right.$$

$$+ \frac{1}{\beta_j} \sum_{k=1}^{j-1} S_k \left(\prod_{p=k}^{j-1} \frac{\lambda_p}{\beta_p} b_{p,p+1} \right) \Big) e^{\beta_{j+1} t} dt$$

$$+ \lambda_j b_{j,j+1} \int \left(\sum_{k=1}^{j-1} \left(\prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\beta_{p+1} - \beta_k} \right) C_k e^{-\beta_k t} \right.$$

$$\left.\left. + C_j e^{-\beta_j t} \right) e^{\beta_{j+1} t} dt \right\} + C_{j+1} e^{-\beta_{j+1} t}$$

$$= \frac{1}{\beta_{j+1}} S_{j+1} + \frac{1}{\beta_{j+1}} \left\{ \frac{\lambda_j}{\beta_j} b_{j,j+1} S_j \right.$$

$$+ \frac{\lambda_j}{\beta_j} b_{j,j+1} \sum_{k=1}^{j-1} S_k \left(\prod_{p=k}^{j-1} \frac{\lambda_p}{\beta_p} b_{p,p+1} \right) \Big\}$$

$$+ \lambda_j b_{j,j+1} \sum_{k=1}^{j-1} \left(\prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\beta_{p+1} - \beta_k} \right) C_k \frac{1}{\beta_{j+1} - \beta_k} e^{-\beta_k t}$$

$$+ \lambda_j b_{j,j+1} C_j \frac{1}{\beta_{j+1} - \beta_j} e^{-\beta_j t} + C_{j+1} e^{-\beta_{j+1} t}$$

$$= \frac{1}{\beta_{j+1}} S_{j+1} + \frac{1}{\beta_{j+1}} \sum_{k=1}^{j} S_k \left(\prod_{p=k}^{j} \frac{\lambda_p}{\beta_p} b_{p,p+1} \right)$$

$$+ \sum_{k=1}^{j} \left(\prod_{p=k}^{j} \frac{\lambda_p b_{p,p+1}}{\beta_{p+1} - \beta_k} \right) C_k e^{-\beta_k t} + C_{j+1} e^{-\beta_{j+1} t}$$

q.e.d

$$C_{j+1} = n_{j+1}(0) - \frac{1}{\beta_{j+1}} S_{j+1} - \frac{1}{\beta_{j+1}} \sum_{k=1}^{j} S_k \left(\prod_{p=k}^{j} \frac{\lambda_p}{\beta_p} b_{p,p+1} \right)$$

$$- \sum_{k=1}^{j} \left(\prod_{p=k}^{j} \frac{\lambda_p b_{p,p+1}}{\beta_{p+1} - \beta_k} \right) C_k$$

which is the same expression as (A-16).

Defining

$$P_{kj} \equiv \prod_{p=k}^{j-1} \frac{\lambda_p}{\beta_p} b_{p,p+1}; \quad k \leq j-1$$

$$P_{jj} \equiv 1$$

$$L_{kj} \equiv \prod_{p=k}^{j-1} \frac{\lambda_p b_{p,p+1}}{\beta_{p+1} - \beta_k}; \quad k \leq j-1$$

$$L_{jj} \equiv 1$$

we obtain the expression (3-37) from (A-15):

$$n_j(t) = \frac{1}{\beta_j} \sum_{k=1}^{j} P_{kj} S_k + \sum_{k=1}^{j} L_{kj} C_k e^{-\beta_k t}$$

Reactor Water Clean Up System. The differential equation (3-41) represents the following series of differential equations

$$\frac{dw_{R1}(t)}{dt} = \varepsilon_1 n_{w1}(t) - \lambda_1 w_{R1}(t) \qquad \text{(A-17)}$$

$$\frac{dw_{R2}(t)}{dt} = \varepsilon_2 n_{w2}(t) + \lambda_1 b_{1,2} w_{R1}(t) - \lambda_2 w_{R2}(t) \qquad \text{(A-18)}$$

.
.
.

$$\frac{dw_{Rj}(t)}{dt} = \varepsilon_j n_{wj}(t) + \lambda_{j-1} b_{j-1,j} w_{Rj-1}(t) - \lambda_j w_{Rj}(t) \qquad \text{(A-19)}$$

Solving first for $w_{R1}(t)$, $w_{R2}(t)$, and $w_{R3}(t)$:

$$w_{R1}(t) = e^{-\int \lambda_1 dt}\left\{\int \varepsilon_1 n_{w1}(t) e^{\int \lambda_1 dt} dt + C_{R1}\right\}$$

$$= e^{-\lambda_1 t}\left\{\varepsilon_1 \int \left(\frac{1}{\beta_1} S_1 + C_1 e^{-\beta_1 t}\right) e^{\lambda_1 t} dt + C_{R1}\right\}$$

$$= \frac{1}{\lambda_1}\frac{\varepsilon_1}{\beta_1} S_1 + \frac{\varepsilon_1}{\lambda_1 - \beta_1} C_1 e^{-\beta_1 t} + C_{R1} e^{-\lambda_1 t} \qquad \text{(A-20)}$$

$$C_{R1} = W_{R1}(0) - \frac{1}{\lambda_1}\frac{\varepsilon_1}{\beta_1} S_1 - \frac{\varepsilon_1}{\lambda_1 - \beta_1} C_1 \qquad \text{(A-21)}$$

$$w_{R2}(t) = e^{-\lambda_2 t}\left\{\int (\varepsilon_2 n_{w2}(t) + \lambda_1 b_{1,2} w_{R1}(t)) e^{\lambda_2 t} dt + C_{R2}\right\}$$

$$= e^{-\lambda_2 t}\left\{\varepsilon_2 \int \left(\frac{1}{\beta_2} S_2 + \frac{1}{\beta_2} P_{1,2} S_1 + L_{1,2} C_1 e^{-\beta_1 t} + C_2 e^{-\beta_2 t}\right) e^{\lambda_2 t} dt\right.$$

$$\left. + \lambda_1 b_{1,2} \int \left(\frac{1}{\lambda_1}\frac{\varepsilon_1}{\beta_1} S_1 + \frac{\varepsilon_1}{\lambda_1 - \beta_1} C_1 e^{-\beta_1 t} + C_{R1} e^{-\lambda_1 t}\right) e^{\lambda_2 t} dt\right\}$$

$$+ C_{R2} e^{-\lambda_2 t}$$

$$= \frac{1}{\lambda_2}\frac{\varepsilon_2}{\beta_2}(S_2 + P_{1,2}S_1) + \frac{\varepsilon_2}{\lambda_2-\beta_1}L_{1,2}C_1 e^{-\beta_1 t}$$

$$+ \frac{\varepsilon_2}{\lambda_2-\beta_2}C_2 e^{-\beta_2 t} + \frac{b_{1,2}}{\lambda_2}\frac{\varepsilon_1}{\beta_1}S_1 + \frac{\varepsilon_1\lambda_1 b_{1,2}}{(\lambda_2-\beta_1)(\lambda_1-\beta_1)}C_1 e^{-\beta_1 t}$$

$$+ \frac{\lambda_1 b_{1,2}}{\lambda_2-\lambda_1}C_{R1}e^{-\lambda_1 t} + C_{R2}e^{-\lambda_2 t}$$

$$= \frac{1}{\lambda_2}\left(B_{1,2}\frac{\varepsilon_1}{\beta_1}S_1 + \frac{\varepsilon_2}{\beta_2}(S_2 + P_{1,2}S_1)\right)$$

$$+ \varepsilon_1\Lambda_{1,2}H_{1,1,2}C_1 e^{-\beta_1 t} + \varepsilon_2(H_{1,2,2}L_{1,2}C_1 e^{-\beta_1 t}$$

$$+ H_{2,2,2}C_2 e^{-\beta_2 t}) + K_{1,2}C_{R1}e^{-\lambda_1 t} + C_{R2}e^{-\lambda_2 t} \qquad \text{(A-22)}$$

$$C_{R2} = w_{R2}(0) - \frac{1}{\lambda_2}\left(B_{1,2}\frac{\varepsilon_1}{\beta_1}S_1 + \frac{\varepsilon_2}{\beta_2}(S_2 + P_{1,2}S_1)\right) - \varepsilon_1\Lambda_{1,2}H_{1,1,2}C_1$$

$$- \varepsilon_2(H_{1,2,2}L_{1,2}C_1 + H_{2,2,2}C_2) - K_{1,2}C_{R1} \qquad \text{(A-23)}$$

In (A-22) and (A-23) the following definitions have been used

$$B_{mj} \equiv \prod_{p=m}^{j-1} b_{p,p+1} \; ; \quad m < j \qquad \text{(A-24)}$$

$$\Lambda_{mj} \equiv \prod_{p=m}^{j-1} \lambda_p b_{p,p+1} \; ; \quad m < j \qquad \text{(A-25)}$$

$$H_{kmj} \equiv \prod_{p=m}^{j} \frac{1}{\lambda_p - \beta_k} \; ; \quad p \geq k \tag{A-26}$$

In this case it is necessary to solve for w_{R3}, too, to obtain a general expression.

$$w_{R3}(t) = e^{-\lambda_3 t}\left\{\int (\varepsilon_3 n_{w3}(t) + \lambda_2 b_{2,3} w_{R2}(t)) e^{\lambda_3 t} dt + C_{R3}\right\}$$

$$= \frac{\varepsilon_3}{\lambda_3}\left\{\frac{1}{\beta_3} S_3 + \frac{1}{\beta_3}\sum_{k=1}^{2} P_{k3} S_k\right\} + \varepsilon_3 \sum_{k=1}^{2} L_{k3} C_k \frac{1}{\lambda_3 - \beta_k} e^{-\beta_k t}$$

$$+ \varepsilon_3 C_3 \frac{1}{\lambda_3 - \beta_3} e^{-\beta_3 t} + \frac{\lambda_2 b_{2,3}}{\lambda_3 \lambda_2}\left\{ B_{1,2} \frac{\varepsilon_1}{\beta_1} S_1 + \frac{\varepsilon_2}{\beta_2}(S_2 + P_{1,2} S_1)\right\}$$

$$+ \frac{\varepsilon_1 \lambda_2 b_{2,3}}{\lambda_3 - \beta_1} \Lambda_{1,2} H_{1,1,2} C_1 e^{-\beta_1 t}$$

$$+ \varepsilon_2 \lambda_2 b_{2,3}\left\{H_{1,2,2} L_{1,2} C_1 \frac{1}{\lambda_3 - \beta_1} e^{-\beta_1 t} + H_{2,2,2} C_2 \frac{1}{\lambda_3 - \beta_2} e^{-\beta_2 t}\right\}$$

$$+ \frac{\lambda_2 b_{2,3}}{\lambda_3 - \lambda_1} K_{1,2} C_{R1} e^{-\lambda_1 t} + \frac{\lambda_2 b_{2,3}}{\lambda_3 - \lambda_2} C_{R2} e^{-\lambda_2 t} + C_{R3} e^{-\lambda_3 t}$$

$$= \frac{1}{\lambda_3}\left\{\frac{\varepsilon_3}{\beta_3}(S_3 + \sum_{k=1}^{2} P_{k3} S_k) + \sum_{m=1}^{2} B_{m3} \frac{\varepsilon_m}{\beta_m}(S_m + \sum_{k=1}^{m-1} P_{km} S_k)\right\}$$

$$+ \varepsilon_3 (H_{3,3,3} C_3 e^{-\beta_3 t} + \sum_{k=1}^{2} H_{k,3,3} L_{k3} C_k e^{-\beta_k t})$$

$$+ \sum_{m=1}^{2} \varepsilon_m \Lambda_{m3} (H_{mm3} C_m e^{-\beta_m t} + \sum_{k=1}^{m-1} H_{km3} L_{km} C_k e^{-\beta_k t})$$

$$+ \sum_{k=1}^{2} K_{k3} C_{Rk} e^{-\lambda_k t} + C_{R3} e^{-\lambda_3 t} \tag{A-27}$$

$$C_{R3} = w_{R3}(0) - \frac{1}{\lambda_3}\left\{\frac{\epsilon_3}{\beta_3}\left(S_3 + \sum_{k=1}^{2} P_{k3}S_k\right) + \sum_{m=1}^{2} B_{m3}\frac{\epsilon_m}{\beta_m}\left(S_m + \sum_{k=1}^{m-1} P_{km}S_k\right)\right\} - \epsilon_3\left(H_{3,3,3}C_3 + \sum_{k=1}^{2} H_{k,3,3}L_{k,3}C_k\right) - \sum_{m=1}^{2}\epsilon_m\Lambda_{m3}\left(H_{mm3}C_m + \sum_{k=1}^{m-1} H_{km3}L_{km}C_k\right) - \sum_{k=1}^{2} K_{k3}C_{Rk} \qquad \text{(A-28)}$$

Note that sums with a lower limit greater than the upper limit must be neglected, and that the following correlations have been used:

$$B_{m,j+1} = B_{mj} \cdot b_{j,j+1} \qquad \text{(A-29)}$$

$$\Lambda_{m,j+1} = \Lambda_{mj} \cdot \lambda_j b_{j,j+1} \qquad \text{(A-30)}$$

$$H_{kmj+1} = H_{kmj} \cdot \frac{1}{\lambda_{j+1} - \beta_k} \qquad \text{(A-31)}$$

It is now postulated that $w_{Rj}(t)$ for $j \geq 3$ follows the expression

$$w_{Rj}(t) = \frac{1}{\lambda_j}\left\{\frac{\epsilon_j}{\beta_j}\left(S_j + \sum_{k=1}^{j-1} P_{kj}S_k\right) + \sum_{m=1}^{j-1} B_{mj}\frac{\epsilon_m}{\beta_m}\left(S_m + \sum_{k=1}^{m-1} P_{km}S_k\right)\right\} + \epsilon_j\left(H_{jjj}C_j e^{-\beta_j t} + \sum_{k=1}^{j-1} H_{kjj}L_{kj}C_k e^{-\beta_k t}\right) + \sum_{m=1}^{j-1}\epsilon_m\Lambda_{mj}\left(H_{mmj}C_m e^{-\beta_m t} + \sum_{k=1}^{m-1} H_{kmj}L_{km}C_k e^{-\beta_k t}\right) + \sum_{k=1}^{j-1} K_{kj}C_{Rk}e^{-\lambda_k t} + C_{Rj}e^{-\lambda_j t} \qquad \text{(A-32)}$$

$$C_{Rj} = w_{Rj}(0) - \frac{1}{\lambda_j}\left\{\frac{\epsilon_j}{\beta_j}\left(S_j + \sum_{k=1}^{j-1} P_{kj}S_k\right) + \sum_{m=1}^{j-1} B_{mj}\frac{\epsilon_m}{\beta_m}\left(S_m + \sum_{k=1}^{m-1} P_{km}S_k\right)\right\} - \epsilon_j\left(H_{jjj}C_j + \sum_{k=1}^{j-1} H_{kjj}L_{kj}C_k\right)$$

$$- \sum_{m=1}^{j-1} \varepsilon_m \Lambda_{mj} (H_{mmj} C_m + \sum_{k=1}^{m-1} H_{kmj} L_{km} C_k) - \sum_{k=1}^{j-1} K_{kj} C_{Rk} \qquad \text{(A-33)}$$

Comparing (A-27) and (A-32) shows that (A-32) is valid for $j = 3$. It will now be proved that $w_{Rj+1}(t)$ follows (A-32) when $w_{Rj}(t)$ does.

$$w_{Rj+1}(t) = e^{-\lambda_{j+1}t}\left\{\int (\varepsilon_{j+1} n_{w,j+1}(t) + \lambda_j b_{j,j+1} w_{Rj}(t)) e^{\lambda_{j+1}t} dt + C_{Rj+1}\right\}$$

$$\equiv I_1 + I_2 + I_3 + I_4 + I_5 + I_6 + C_{R,j+1} e^{-\lambda_{j+1}t} \qquad \text{(A-34)}$$

For clarity, the integral has been split up into 6 integrals, which are defined and evaluated below:

$$I_1 \equiv e^{-\lambda_{j+1}t} \varepsilon_{j+1} \int (\frac{1}{\beta_{j+1}} S_{j+1} + \frac{1}{\beta_{j+1}} \sum_{k=1}^{j} P_{k,j+1} S_k) e^{\lambda_{j+1}t} dt$$

$$= \frac{1}{\lambda_{j+1}} \frac{\varepsilon_{j+1}}{\beta_{j+1}} (S_{j+1} + \sum_{k=1}^{j} P_{k,j+1} S_k) \qquad \text{(A-35)}$$

$$I_2 \equiv e^{-\lambda_{j+1}t} \varepsilon_{j+1} \int (C_{j+1} e^{-\beta_{j+1}t} + \sum_{k=1}^{j} L_{k,j+1} C_k e^{-\beta_k t}) e^{\lambda_{j+1}t} dt$$

$$= \frac{\varepsilon_{j+1}}{\lambda_{j+1} - \beta_{j+1}} C_{j+1} e^{-\beta_{j+1}t} + \varepsilon_{j+1} \sum_{k=1}^{j} L_{k,j+1} C_k \frac{1}{\lambda_{j+1} - \beta_k} e^{-\beta_k t}$$

$$= \varepsilon_{j+1} H_{j+1,j+1,j+1} C_{j+1} e^{-\beta_{j+1}t} + \varepsilon_{j+1} \sum_{k=1}^{j} H_{k,j+1,j+1} L_{k,j+1} C_k e^{-\beta_k t} \qquad \text{(A-36)}$$

$$I_3 \equiv e^{-\lambda_{j+1}t} \lambda_j b_{j,j+1} \frac{1}{\lambda_j} \int \left\{\frac{\varepsilon_j}{\beta_j} (S_j + \sum_{k=1}^{j-1} P_{kj} S_k) + \sum_{m=1}^{j-1} B_{mj} \frac{\varepsilon_m}{\beta_m} (S_m + \sum_{k=1}^{m-1} P_{km} S_k)\right\} e^{\lambda_{j+1}t} dt$$

$$= \frac{b_{j,j+1}}{\lambda_{j+1}}\left\{ \frac{\epsilon_j}{\beta_j}(S_j + \sum_{k=1}^{j-1} P_{kj}S_k) + \sum_{m=1}^{j-1} B_{mj} \frac{\epsilon_m}{\beta_m}(S_m + \sum_{k=1}^{m-1} P_{km}S_k)\right\}$$

$$= \frac{1}{\lambda_{j+1}} \sum_{m=1}^{j} B_{m,j+1} \frac{\epsilon_m}{\beta_m}(S_m + \sum_{k=1}^{m-1} P_{km}S_k) \qquad \text{(A-37)}$$

where the correlation (A-29) and the definition of $B_{j,j+1}$ have been utilised.

$$I_4 \equiv e^{-\lambda_{j+1}t} \lambda_j b_{j,j+1} \epsilon_j \int \left\{ H_{jjj} C_j e^{-\beta_j t} + \sum_{k=1}^{j-1} H_{kjj} L_{kj} C_k e^{-\beta_k t} \right\} e^{\lambda_{j+1}t} dt$$

$$= \frac{\lambda_j b_{j,j+1} \epsilon_j}{\lambda_{j+1} - \beta_j} H_{jjj} C_j e^{-\beta_j t} + \lambda_j b_{j,j+1} \epsilon_j \sum_{k=1}^{j-1} H_{kjj} L_{kj} C_k \frac{1}{\lambda_{j+1} - \beta_k} e^{-\beta_k t}$$

$$= \epsilon_j \Lambda_{j,j+1} (H_{jjj+1} C_j e^{-\beta_j t} + \sum_{k=1}^{j-1} H_{kjj+1} L_{kj} C_k e^{-\beta_k t}) \qquad \text{(A-38)}$$

Here (A-25) and (A-31) have been used.

$$I_5 \equiv e^{-\lambda_{j+1}t} \lambda_j b_{j,j+1} \int \left\{ \sum_{m=1}^{j-1} \epsilon_m \Lambda_{mj} \left(H_{mmj} C_m e^{-\beta_m t} \right.\right.$$

$$\left.\left. + \sum_{k=1}^{m-1} H_{kmj} L_{km} C_k e^{-\beta_k t} \right) \right\} e^{\lambda_{j+1}t} dt$$

$$= \sum_{m=1}^{j-1} \epsilon_m \Lambda_{m,j+1} \left(H_{mmj+1} C_m e^{-\beta_m t} + \sum_{k=1}^{m-1} H_{kmj+1} L_{km} C_k e^{-\beta_k t} \right) \qquad \text{(A-39)}$$

$$I_6 \equiv e^{-\lambda_{j+1}t} \lambda_j b_{j,j+1} \int \left\{ \sum_{k=1}^{j-1} K_{kj} C_{Rk} e^{-\lambda_k t} + C_{Rj} e^{-\lambda_j t} \right\} e^{\lambda_{j+1}t} dt$$

$$= \sum_{k=1}^{j-1} K_{kj} \frac{\lambda_j b_{j,j+1}}{\lambda_{j+1} - \lambda_k} C_{Rk} e^{-\lambda_k t} + \frac{\lambda_j b_{j,j+1}}{\lambda_{j+1} - \lambda_j} C_{Rj} e^{-\lambda_j t}$$

$$= \sum_{k=1}^{j-1} K_{kj+1} C_{Rk} e^{-\lambda_k t} + K_{j,j+1} C_{Rj} e^{-\lambda_j t}$$

$$= \sum_{k=1}^{j} K_{k,j+1} C_{Rk} e^{-\lambda_k t} \qquad \text{(A-40)}$$

$$I_1 + I_3 = \frac{1}{\lambda_{j+1}} \left\{ \frac{\epsilon_{j+1}}{\beta_{j+1}} (S_{j+1} + \sum_{k=1}^{j} P_{k,j+1} S_k) + \sum_{m=1}^{j} B_{m,j+1} \frac{\epsilon_m}{\beta_m} (S_m + \sum_{k=1}^{m-1} P_{km} S_k) \right\} \qquad \text{(A-41)}$$

$$I_4 + I_5 = \sum_{m=1}^{j} \epsilon_m \Lambda_{m,j+1} (H_{mmj+1} C_m e^{-\beta_m t} + \sum_{k=1}^{m-1} H_{kmj+1} L_{km} C_k e^{-\beta_k t}) \qquad \text{(A-42)}$$

$$w_{R,j+1}(t) = (I_1 + I_3) + I_2 + (I_4 + I_5) + I_6 + C_{R,j+1} e^{-\lambda_{j+1} t} \qquad \text{(A-43)}$$

Comparing term by term with (A-32) reveals that the expression (A-43) is identical to (A-32) when replacing index j with j+1, which was to be proved.

Now, the expression for $w_{Rj}(t)$ can be made slightly more compact by extending the definitions (A-24) and (A-25) to cover the cases m = j, as was done in (3-43) and (3-44). Hereby expression (3-42) is obtained. Furthermore it is easily verified that cases j = 1 and j = 2 fit (3-42) as well.

APPENDIX B

Derivation of Inventories During Shutdown Assuming Logarithmic Corrosion Kinetics

Assuming logarithmic corrosion kinetics, the emission rate from the construction material of a component in the reactor system can be expressed as follows.

For nuclide no. 1, the inactive target nuclide:

$$Q_{i1}(t) = \frac{N_{Av}}{M_{A,s}} a_j^s \sum_m A_{m,i} p_m^s \frac{K_{\ell,m}\, a_{\ell,m}}{a_{\ell,m}^{t+1}} , \qquad \text{(B-1)}$$

and for the activation product nuclide

$$Q_{i2}(t) = Q_{i1}(t) \cdot F_i(t_o,t_1) e^{-\lambda_2(t-t_o)} \qquad \text{(B-2)}$$

cf. section 4.1.1.1. for a definition of the parameters. The emission rates for the reactor water system as a whole are

$$Q_{w1}(t) = \sum_{RWC} Q_{i1}(t) = \sum_{RWC} \frac{N_{Av}}{M_{A,s}} a_j^s \sum_m A_{m,i} p_m^s \frac{K_{\ell,m}\, a_{\ell,m}}{a_{\ell,m}^{t+1}}$$

$$\equiv \sum_{RWC} \sum_m D_{m,i} \frac{a_m}{a_m^{t+1}} \qquad \text{(B-3)}$$

where the constants that are unimportant in this context have been combined in a new set of constants, $D_{m,i}$, and $a_{\ell,m}$ has been changed to a_m.

$$Q_{w2}(t) = \sum_{RWC} Q_{i2}(t) = \sum_{RWC} Q_{i1}(t) F_i(t_o,t_1) e^{-\lambda_2(t-t_o)}$$

$$= \sum_{RWC} \sum_m D_{m,i} F_i(t_o,t_1) \frac{a_m}{a_m^{t+1}} e^{-\lambda_2(t-t_o)} \qquad \text{(B-4)}$$

With the emission rates expressed by (B-3) and (B-4), the inventories are now evaluated for the reactor water and RWCU system.

Reactor Water System

Non-Radioactive Nuclide, $\varepsilon_1 > 0$

$$n_{w1}(t) = e^{-\varepsilon_1 t}\left\{\int Q_{w1}(t) e^{\varepsilon_1 t} dt + C_1\right\}$$

$$= e^{-\varepsilon_1 t}\left\{\sum_{RWC}\sum_{m} D_{m,i}\int \frac{a_m}{a_m t+1} e^{\varepsilon_1 t} dt + C_1\right\} \qquad \text{(B-5)}$$

$$\int \frac{a_m}{a_m t+1} e^{\varepsilon_1 t} dt = \int \frac{1}{x} e^{\varepsilon_1 \left(\frac{x-1}{a_m}\right)} dx = e^{-b}\int \frac{1}{x} e^{bx} dx$$

$$= e^{-b}\left\{ \ln x + \frac{bx}{1!} + \frac{b^2x^2}{2\cdot 2!} + \frac{b^3x^3}{3\cdot 3!} + \cdots\right\}$$

$$= e^{-b}\left\{ \ln x + \sum_{n=1}^{\infty} \frac{(bx)^n}{n\cdot n!} \right\}$$

$$= e^{-\varepsilon_1/a_m}\left\{ \ln(a_m t+1) + \sum_{n=1}^{\infty} \frac{(\varepsilon_1 t+\varepsilon_1/a_m)^n}{n\cdot n!} \right\} \qquad \text{(B-6)}$$

where the substitutions $x \equiv a_m t+1$ and $b \equiv \varepsilon_1/a_m$ have been used. Insertion of (B-6) into (B-5) gives

$$n_{w1}(t) = \sum_{RWC}\sum_{m} D_{m,i} e^{-(\varepsilon_1 t+\varepsilon_1/a_m)}\left\{\ln(a_m t+1) + \sum_{n=1}^{\infty} \frac{(\varepsilon_1 t+\varepsilon_1/a_m)^n}{n\cdot n!}\right\}$$

$$+ C_1 e^{-\varepsilon_1 t} \qquad \text{(B-7)}$$

$$C_1 = n_{w1}(t_o) e^{\varepsilon_1 t_o} - \sum_{RWC}\sum_{m} D_{m,i} e^{-\varepsilon_1/a_m}\left\{ \ln(a_m t_o+1)\right.$$

$$\left. + \sum_{n=1}^{\infty} \frac{(\varepsilon_1 t_o+\varepsilon_1/a_m)^n}{n\cdot n!} \right\} \qquad \text{(B-8)}$$

The infinite series $\sum_{n=1}^{\infty} \frac{(\varepsilon_1 t_o+\varepsilon_1/a_m)^n}{n\cdot n!}$ is convergent for all

values of t_o. This can be proved by showing that the n'th and n+1'st term fulfil the condition

$$\left|\frac{u_{n+1}}{u_n}\right| < 1 \text{ for } n > N, \text{ where N is a finite number.}$$

$$\frac{(\varepsilon_1 t_o+\varepsilon_1/a_m)^{n+1}}{(n+1)(n+1)!} \Big/ \frac{(\varepsilon_1 t_o+\varepsilon_1/a_m)^{n}}{n\cdot n!} = \frac{(\varepsilon_1 t_o+\varepsilon_1/a_m)\cdot n}{(n+1)^2}$$

$$< \frac{\varepsilon_1 t_o+\varepsilon_1/a_m}{n} < 1 \text{ for } n > N = \varepsilon_1 t_o+\varepsilon_1/a_m$$

q.e.d.

Non-Radioactive Nuclide, $\varepsilon_1=0$

$$n_{w1}(t) = n_{w1}(t_o) + \int_{t_o}^{t} Q_{w1}(\tau)d\tau$$

$$= n_{w1}(t_o) + \sum_{RWC}\sum_{m} D_{m,i} \int_{t_o}^{t} \frac{a_m}{a_m\tau+1} d\tau$$

$$= n_{w1}(t_o) + \sum_{RWC}\sum_{m} D_{m,i} \ln \frac{a_m t+1}{a_m t_o+1} \qquad \text{(B-9)}$$

Activation Product Nuclide

$$n_{w2}(t) = e^{-\beta_2 t}\left\{\int Q_{w2}(t)e^{\beta_2 t}dt + C_2\right\}$$

$$= e^{-\beta_2 t}\left\{\sum_{RWC}\sum_{m} D_{m,i}F_i(t_o,t_1)\int \frac{a_m}{a_m t+1} e^{-\lambda_2(t-t_o)}e^{\beta_2 t}dt + C_2\right\}$$

$$= e^{-\beta_2 t}\left\{\sum_{RWC}\sum_{m} D_{m,i}F_i(t_o,t_1)e^{\lambda_2 t_o}\int \frac{a_m}{a_m t+1} e^{\varepsilon_2 t}\, dt + C_2\right\}$$

(B-10)

The integral in (B-10) is the same as that in (B-5), when

replacing ϵ_2 by ϵ_1. So the solution for $n_{w2}(t)$ becomes

$$n_{w2}(t) = e^{-(\beta_2 t-\lambda_2 t_o)} \sum_{RWC} \sum_m D_{m,i} F_i(t_o,t_1) e^{-\epsilon_2/a_m} \left\{ \ln(a_m t+1) + \sum_{n=1}^{\infty} \frac{(\epsilon_2 t+\epsilon_2/a_m)^n}{n \cdot n!} \right\} + C_2 e^{-\beta_2 t} \qquad \text{(B-11)}$$

$$C_2 = n_{w2}(t_o) e^{\beta_2 t_o} - e^{\lambda_2 t_o} \sum_{RWC} \sum_m D_{m,i} F_i(t_o,t_1) e^{-\epsilon_2/a_m} \left\{ \ln(a_m t_o+1) + \sum_{n=1}^{\infty} \frac{(\epsilon_2 t_o+\epsilon_2/a_m)^n}{n \cdot n!} \right\} \qquad \text{(B-12)}$$

Expressions (B-11) and (B-12) will apply to both cases $\epsilon_2=0$ and $\epsilon_2>0$.

Reactor Water Clean Up System

Non-Radioactive Nuclide

$$w_{R1}(t) = w_{R1}(t_o) + \int_{t_o}^{t} \epsilon_1 n_{w1}(\tau) d\tau$$

$$= w_{R1}(t_o) + \epsilon_1 \int_{t_o}^{t} \sum_{RWC} \sum_m D_{m,i} e^{-(\epsilon_1 \tau+\epsilon_1/a_m)} \left\{ \ln(a_m \tau+1) + \sum_{n=1}^{\infty} \frac{(\epsilon_1 \tau+\epsilon_1/a_m)^n}{n \cdot n!} \right\} d\tau + \epsilon_1 C_1 \int_{t_o}^{t} e^{-\epsilon_1 \tau} d\tau$$

$$= w_{R1}(t_o) + \sum_{RWC} \sum_m D_{m,i} \int_{Y_o}^{Y} e^{-y} \left\{ \ln \frac{a_m}{\epsilon_1} + \ln y + \sum_{n=1}^{\infty} \frac{y^n}{n \cdot n!} \right\} dy + \epsilon_1 C_1 \int_{t_o}^{t} e^{-\epsilon_1 \tau} d\tau \qquad \text{(B-13)}$$

where the following substitutions have been introduced:

$$y \equiv \epsilon_1 \tau+\epsilon_1/a_m, \quad dy \equiv \epsilon_1 d\tau,$$

$Y \equiv c_1 t + c_1/a_m$, and $Y_o \equiv c_1 t_o + c_1/a_m$.

The integrals in (B-13) are now split up into four and are evaluated one by one below.

$$\int_{Y_o}^{Y} e^{-y} \ln \frac{a_m}{c_1}\, dy = (e^{-Y_o} - e^{-Y}) \ln \frac{a_m}{c_1} \tag{B-14}$$

$$\int_{Y_o}^{Y} e^{-y} \ln y\, dy = \left[-e^{-y} \ln y\right]_{Y_o}^{Y} + \int_{Y_o}^{Y} \frac{e^{-y}}{y}\, dy$$

$$= e^{-Y_o} \ln Y_o - e^{-Y} \ln Y + \left[\ln y + \sum_{n=1}^{\infty} \frac{(-y)^n}{n \cdot n!} \right]_{Y_o}^{Y}$$

$$= e^{-Y_o} \ln Y_o - e^{-Y} \ln Y + \ln \frac{Y}{Y_o} + \sum_{n=1}^{\infty} \frac{(-Y)^n - (-Y_o)^n}{n \cdot n!} \tag{B-15}$$

$$\int_{Y_o}^{Y} e^{-y} \sum_{n=1}^{\infty} \frac{y^n}{n \cdot n!}\, dy = \sum_{n=1}^{\infty} \frac{1}{n \cdot n!} \int_{Y_o}^{Y} e^{-y} y^n dy$$

$$= \sum_{n=1}^{\infty} \frac{1}{n \cdot n!} \left[-e^{-y} \sum_{r=0}^{n} \frac{n! y^{n-r}}{(n-r)!} \right]_{Y_o}^{Y}$$

$$= \sum_{n=1}^{\infty} \sum_{r=0}^{n} \frac{e^{-Y_o} Y_o^{n-r} - e^{-Y} Y^{n-r}}{n \cdot (n-r)!} \tag{B-16}$$

$$\int_{t_o}^{t} e^{-c_1 \tau}\, d\tau = \frac{1}{c_1} (e^{-c_1 t_o} - e^{-c_1 t}) = \frac{1}{c_1} (e^{-Y_o} - e^{-Y}) e^{c_1/a_m} \tag{B-17}$$

Inserting (B-14) - (B-17) into (B-13) gives the voluminous expression

$$w_{R1}(t) = w_{R1}(t_o) + \sum_{RWC} \sum_{m} D_{mi} (e^{-Y_o} - e^{-Y}) \ln \frac{a_m}{c_1}$$

$$+ \sum_{RWC} \sum_{m} D_{mi} \left\{ e^{-Y_o} \ln Y_o - e^{-Y} \ln Y + \ln \frac{Y}{Y_o} + \sum_{n=1}^{\infty} \frac{(-Y)^n - (-Y_o)^n}{n \cdot n!} \right\}$$

$$+ \sum_{RMC} \sum_{m} D_{mi} \sum_{n=1}^{\infty} \sum_{r=0}^{n} \frac{e^{-Y_o} Y_o^{n-r} - e^{-Y_Y} Y_Y^{n-r}}{n!(n-r)!}$$

$$+ C_1 (e^{-Y_o} - e^{-Y}) e^{\epsilon_1/a_m} \qquad \text{(B-18)}$$

Activation Product Nuclide

$$w_{R2}(t) = e^{-\lambda_2 t} \left\{ \int \epsilon_2 n_{w2}(t) e^{\lambda_2 t} dt + C_R \right\}$$

$$= e^{-\lambda_2 t} \int \epsilon_2 e^{-(\beta_2 t - \lambda_2 t_o)} \sum_{RMC} \sum_{m} D_{m,i} F_i(t_o, t_1) e^{-\epsilon_2/a_m} \left\{ \ln(a_m t + 1) \right.$$

$$\left. + \sum_{n=1}^{\infty} \frac{(\epsilon_2 t + \epsilon_2/a_m)^n}{n \cdot n!} \right\} e^{\lambda_2 t} dt + e^{-\lambda_2 t} \int \epsilon_2 C_2 e^{-\beta_2 t} e^{\lambda_2 t} dt + C_R e^{-\lambda_2 t}$$

$$= e^{-\lambda_2 (t - t_o)} \epsilon_2 \sum_{RMC} \sum_{m} D_{m,i} F_i(t_o, t_1) e^{-\epsilon_2/a_m} \int \left\{ \ln(a_m t + 1) \right.$$

$$\left. + \sum_{n=1}^{\infty} \frac{(\epsilon_2 t + \epsilon_2/a_m)^n}{n \cdot n!} \right\} e^{-\epsilon_2 t} dt - C_2 e^{-\beta_2 t} + C_R e^{-\lambda_2 t}$$

(B-19)

As is easily verified, the integral in (B-19) occurs in (B-13) as well, and has been evaluated in (B-14) - (B-16). So

$$w_{R2}(t) = e^{-\lambda_2 (t - t_o)} \epsilon_2 \sum_{RMC} \sum_{m} D_{m,i} F_i(t_o, t_1) \left\{ -e^{-(\epsilon_2 t + \epsilon_2/a_m)} \ln(a_m t + 1) \right.$$

$$+ \ln(\epsilon_2 t + \epsilon_2/a_m) + \sum_{n=1}^{\infty} \frac{(-\epsilon_2 t - \epsilon_2/a_m)^n}{n \cdot n!}$$

$$\left. - \sum_{n=1}^{\infty} \sum_{r=0}^{n} e^{-(\epsilon_2 t + \epsilon_2/a_m)} \frac{(\epsilon_2 t + \epsilon_2/a_m)^{n-r}}{n \cdot (n-r)!} \right\}$$

$$- C_2 e^{-\beta_2 t} + C_R e^{-\lambda_2 t} \qquad \text{(B-20)}$$

$$C_R = w_{R2}(t_o)e^{\lambda_2 t_o - e^{\lambda_2 t_o}\varepsilon_2} \sum_{RWC} \sum_m D_{mi} F_i(t_o, t_1) \times$$

$$\left\{ -e^{-(\varepsilon_2 t_o + \varepsilon_2/a_m)} \ln(a_m t_o + 1) + \ln(\varepsilon_2 t_o + \varepsilon_2/a_m) \right.$$

$$\left. + \sum_{n=1}^{\infty} \frac{(-\varepsilon_2 t_o - \varepsilon_2/a_m)^n}{n \cdot n!} - \sum_{n=1}^{\infty} \sum_{r=0}^{n} e^{-(\varepsilon_2 t_o + \varepsilon_2/a_m)} \frac{(\varepsilon_2 t_o + \varepsilon_2/a_m)^{n-r}}{n \cdot (n-r)!} \right\}$$

$$+ C_2 e^{-\varepsilon_2 t_o} \qquad \text{(B-21)}$$

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